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Radiological Health

# PROCEDURES FOR DETERMINATION OF STABLE ELEMENTS AND RADIONUCLIDES IN ENVIRONMENTAL SAMPLES

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Public Health Service

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# PROCEDURES FOR DETERMINATION OF STABLE ELEMENTS AND RADIONUCLIDES IN ENVIRONMENTAL SAMPLES

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## PREFACE

The Division of Radiological Health, Public Health Service, has three regional laboratories engaged primarily in environmental radiological surveillance. Procedures for the analysis of environmental samples have been developed at these laboratories through experience and experiment and have been tested and proven by the successful analyses of hundreds of routine and special environmental samples.

This manual is a compilation of the procedures presently used at the Southeastern Radiological Health Laboratory, Montgomery, Ala. for the determination of stable elements and radionuclides in environmental samples. It should be noted that these procedures are intended for use in the processing of relatively large numbers of samples in the shortest possible time for environmental radiological surveillance and, therefore, in some cases represent a compromise between precise analytical determination and adequate determination for surveillance purposes. It is intended that these procedures be revised and additional methods and information included as the need arises.

The use in this manual of trade names of commercially available products is only to indicate that the particular product is being used in the laboratory. Such use of the name or the product does not constitute an endorsement of the product by the Public Health Service and does not imply that there are no other suitable products available commercially.

## ABSTRACT

This is a manual of methods used at the Public Health Service's Southeastern Radiological Health Laboratory for the determination of stable elements and radionuclides in environmental samples. The procedures are intended for use in the processing of relatively large numbers of samples in a minimum of time for environmental radiological surveillance. The manual is divided into sections on: milk; food, biota, silt, and soil; water; and counting procedures. Appendixes cover preparation of reagents and tables of selected radionuclide decay and ingrowth factors.

## ACKNOWLEDGMENTS

The editors gratefully acknowledge the assistance in the preparation of this manual of the following staff members of the SERHL:

Mary W. Chesnutt
Rachael H. Gorrie
Georgia Lindsey
Estie W. Pepper
Charles R. Phillips
Emmett F. Reeder
Myrtice Smith
Ann B. Strong

Former members of the staff contributing to the initial writing of the procedures used at the SERHL include:

Dr. Bernd Kahn
Daniel F. Cahill
Ronald A. Caton
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# SECTION I. MILK

## A. Strontium-90

### Principle of Method

Milk, with added citrate solution containing yttrium, strontium, and barium carriers, is passed successively through cation- and anion-exchange resin columns. Strontium, barium, and calcium are adsorbed on the cation-exchange resin, and the yttrium carrier, with the yttrium-90 daughter of strontium-90, is retained on the anion-exchange resin. The yttrium is eluted from the anion resin with hydrochloric acid and precipitated as the oxalate. Lanthanum-140 may be a contaminant. To remove the lanthanum contaminant, yttrium oxalate is dissolved in concentrated nitric acid and yttrium extracted from the solution into an equal volume of preequilibrated tributyl phosphate. The lanthanum-140 remains in the concentrated nitric acid to be discarded. Yttrium is reextracted from the organic phase with dilute nitric acid and precipitated as the oxalate. The precipitate is weighed to determine recovery of yttrium carrier, then counted. Calcium, strontium, and barium are eluted from the cation-exchange resin with sodium chloride solution. An aliquot of this eluate is taken for the determination of stable calcium. The remaining eluate is used to determine barium-140 and "total radiostrontium." After washing the anion-exchange resin with additional hydrochloric acid, both columns are washed with distilled water and the resins are again ready for use. This method may be used for the analysis of milk containing fresh fission products as well as milk which contains only long-lived fission products.

### Reagents*

Ammonium hydroxide, 15N
Anion-exchange resin, Dowex-1-X8 ($Cl^-$ form) (50-100 mesh)
Barium carrier, 20 mg $Ba^{+2}$/ml
Cation-exchange resin, Dowex-50W-X8 ($Na^+$ form) (50-100 mesh)
Citrate solution, 3N (pH 6.5)
Diethyl ether
Ethyl alcohol, 95%

*See appendix A.



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Hydrochloric acid, 6N, 2N
Nitric acid, 16N, 14N, 0.1N
Oxalic acid, 2N
Sodium chloride, 4N
Strontium carrier, 20 mg $Sr^{+2}$/ml
Tributyl phosphate (TBP), preequilibrated
Yttrium carrier, 10 mg $Y^{+3}$/ml

## Apparatus

Ion-exchange system (fig. 1-1): Connect glass reservoir (graduated, 1-liter separatory funnel) and two glass columns (with extra coarse fritted glass discs in bottom) one above the other, for gravity flow. Fill upper (5-cm diameter) column with 170 ml Dowex-50W resin, and lower (1.9-cm diameter) column with 15 ml Dowex-1 resin by pouring resin into water-filled columns.
Analytical balance
Millipore filtering apparatus
Millipore #OH membrane-filter (1.5-$\mu$ pore size), 2.5-cm diameter
pH meter
Separatory funnels, 1 liter, 500 ml, 125 ml, 60 ml
Stainless steel planchets, 2-in. diameter

## Procedure

1. Place 1 liter of milk (note a) into the graduated reservoir. Pipette 1.0 ml each of yttrium, strontium, and barium carriers into 10 ml of citrate solution, swirl to mix and dissolve barium citrate. Transfer quantitatively to milk with 5 ml of distilled water and mix well.

2. Open stopcocks of reservoir, anion column, and cation column, in that order. Control flow rate at 10 ml per minute (check occasionally by collecting effluent for 1 minute in graduated cylinder) with anion column stopcock. Stop flow when just enough milk remains in columns to cover resin. Discard effluent milk. RECORD MIDPOINT OF ELUTION TIME AS START OF YTTRIUM-90 DECAY.

3. Replace milk reservoir with a separatory funnel containing 300 ml of warm distilled water and wash columns with water (flow rate of approximately 10 ml per minute) to displace milk. Stop flow when just enough water remains in columns to cover resin. Discard effluent water.

4. Separate columns: In order to collect eluate for "total radiostrontium", barium, and calcium determinations, and to regenerate

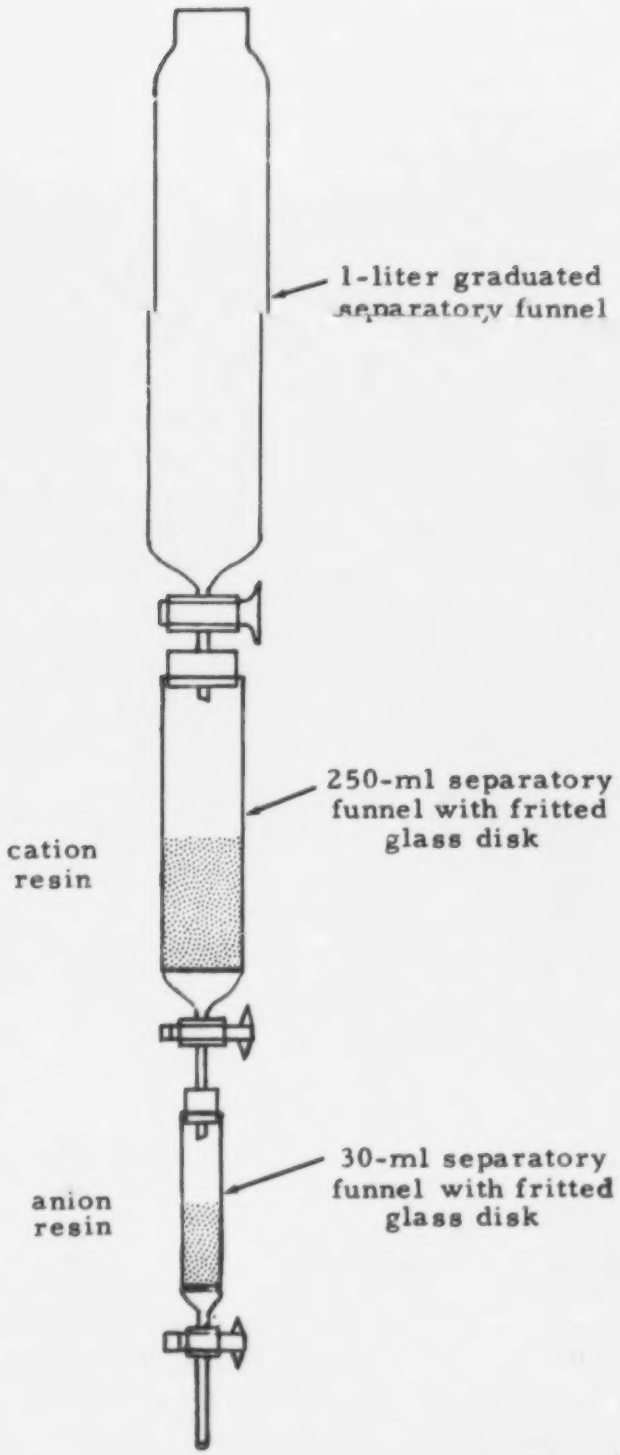
1-liter graduated separatory funnel
250-ml separatory funnel with fritted glass disk
cation resin
30-ml separatory funnel with fritted glass disk
anion resin

FIGURE 1-1. Ion-Exchange System

the cation column for subsequent use, follow step 4a. For strontium-90 determination proceed with step 5.

4a. To the top of cation column, attach a 1-liter separatory funnel containing 1 liter of 4N NaCl. Allow the NaCl solution to flow at 10 ml per minute to elute alkali metal and alkaline earth ions and also to recharge the column. Leave enough eluent to cover resin to a depth of 2 cm. Collect eluate until the total volume is 1 liter. For analysis of strontium-89 and barium-140, continue with step 6 of section I, part B. For calcium determination, continue with step 2 of section I, part C. To wash the column for reuse, place 500 ml of distilled water in a separatory funnel and let flow into another beaker at 10 ml per minute to remove excess NaCl from the resin (note b). Discard this washing.

5. Attach the separatory funnel containing 100 ml of 2N HCl to the top of the anion column. Let acid flow at 2 ml per minute. Discard effluent until pH drops to 2 (after 10 to 20 ml). Collect the next 10 ml of eluate, stop flow of acid and remove separatory funnel. Stir the anion resin thoroughly with a glass stirring rod. Wash the stirring rod with a small amount of 2N HCl and add this acid to the resin column. Reattach the separatory funnel and continue eluting with 2N HCl. Collect the next 25 ml of eluate to a total volume of 35 ml. Continue processing this eluate for strontium-90 analysis at step 7 below (note c).

6. Allow remaining 2N Hcl to flow at 10 ml per minute to wash and recharge the column. Discard effluent. Wash resin with 100 ml of distilled water and discard effluent water (note b).

7. To precipitate yttrium oxalate from the 35 ml of anion eluate, add 5 ml of 2N oxalic acid, and adjust to pH 1.5 with approximately 2.5 ml of 15N $NH_4OH$ (using pH meter). Stir, heat to near boiling, cool in an ice bath approximately 20 minutes, centrifuge, and discard supernate.

8. If fresh fission products are present in the sample, follow steps 9-19. If fresh fission products are <u>not</u> present, the lanthanum extraction procedure (steps 9-14) may be omitted. In this case follow step 8a and proceed to step 15.

8a. Wash precipitate with 10 ml of hot distilled water, centrifuge, decant, and discard supernate. Dissolve precipitate in 1 ml of 6N HCl and 15 ml of hot distilled water. If insoluble material remains, centrifuge and transfer supernate to another 40-ml centrifuge tube. Continue working with supernate in step 15.

9. Add 10 ml of 16N $HNO_3$ to dissolve the precipitate. Transfer from centrifuge tube to a 60-ml separatory funnel. Wash centrifuge tube with 10 ml of preequilibrated TBP and add this TBP to the separatory funnel.

10. Equilibrate by shaking for approximately 2-3 minutes; allow phases to separate. Drain acid phase away and discard.

11. Add 15 ml of 14N $HNO_3$ to the separatory funnel and equilibrate again for approximately 2-3 minutes; allow phases to separate. Drain acid phase away and discard.

12. Repeat step 11.

13. Remove yttrium from the organic phase by adding 15 ml of distilled water and equilibrating for approximately 2-3 minutes. Allow phases to separate and drain aqueous phase into 40-ml centrifuge tube.

14. Repeat step 13 using 15 ml of 0.1N $HNO_3$ instead of distilled water and drain acid phase into same 40-ml centrifuge tube as in step 13.

15. To precipitate yttrium oxalate, add 5 ml of 2N oxalic acid and adjust to pH 1.5 with approximately 2 ml of 15N $NH_4OH$ (using pH meter). Stir and cool in an ice bath approximately 20 minutes. Centrifuge and discard supernate.

16. Weigh a stainless steel planchet and 2.5-cm diameter membrane filter. Place membrane filter into filtering apparatus.

17. Take up precipitate in 10 ml of cold water and transfer to filtering apparatus. Wash with one 10-ml portion of cold distilled water, using the water to wash out centrifuge tube. Dry precipitate with three 20-ml portions of 95% ethyl alcohol and three 10-ml portions of diethyl ether. Continue suction for 2 minutes.

18. Transfer membrane filter with precipitate to stainless steel planchet and weigh after a delay of 10-15 minutes. Calculate the recovery of yttrium carrier.

19. Count the yttrium oxalate precipitate in a low-background beta counter (note d). Count again after 3 days to confirm absence of contamination of yttrium-90 by other radionuclides.

Notes

a. Milk must be reasonably homogeneous, and should be preserved with formaldehyde and stored under refrigeration (32° to 34° F) for 2 weeks. Large amounts of separated cream or solids will interfere with or prevent flow of milk through resin columns. If clogging occurs, stirring of the resin may increase the flow. For procedure for nonhomogeneous milk, see section I, part D.

b. If resins clog with extended use, backwashing or slurrying of the resin with distilled water will remove trapped milk particulates.

c. Prior to the passage of milk over the column, 10 ml of tincture of merthiolate containing eosin dye can be thoroughly mixed with the anion resin to act as an indicator when eluting with 2 N HCl. This aids in determining the point at which the pH drops to 2. However, universal pH paper is used to determine the exact point at which the pH change occurs.

d. Care must be exercised that membrane filter remains centered in planchet to avoid any geometric effects during counting.

Calculation

Strontium-90: $$pc/l = \frac{(\text{net cpm})}{(A)(B)(C)(D)(E)}$$

where (A) = recovery of yttrium carrier,

(B) = counting efficiency in $\frac{cpm}{pc}$ for yttrium 90 as yttrium oxalate mounted on 2.5-cm membrane filter

(since 1 pc = 2.22 dpm, $\frac{cpm}{pc} = 2.22 \frac{cpm}{dpm}$),

(C) = sample volume in liters,

(D) = correction factor for yttrium-90 decay ($e^{-\lambda t}$), where t is the time from midpoint of the elution time to the time of counting,*

(E) = correction factor for the degree of equilibrium attained during the yttrium-90 ingrowth period ($1-e^{-\lambda t}$), where t is the time from the collection of the milk sample to the time of passage through the column.**

*Obtained from table 1, "Y-90 Decay and Ingrowth Factors", appendix B.
**Obtained from table 2, "Y-90 Ingrowth Factors", appendix B.

## B. Strontium-89 and Barium-140

### Principle of Method

Milk, with added citrate solution containing yttrium, strontium, and barium carriers, is passed successively through cation- and anion-exchange resin columns. Strontium, barium, and calcium are adsorbed on the cation-exchange resin, then eluted with sodium chloride solution. Following dilution of the eluate the alkaline earths are precipitated as the carbonates and converted to the nitrates, precipitating strontium and barium nitrate. The nitrate precipitate is dissolved and barium is precipitated as the chromate, purified as the chloride, and then counted to determine barium-140. Strontium is precipitated from the supernate as the nitrate, dissolved in deionized distilled water, and reprecipitated as the nitrate. The nitrate is dissolved and precipitated as strontium oxalate. The oxalate precipitate is filtered, weighed to determine strontium carrier recovery, and counted for "total radiostrontium". The strontium-89 activity is calculated from the difference between the count for "total radiostrontium" and the count due to the value previously obtained for strontium-90 activity.

### Reagents*

Ammonium acetate buffer, pH 5
Ammonium hydroxide, 15N
Ammonium oxalate, 1N
Anion-exchange resin, Dowex-1-X8 ($Cl^-$ form) (50-100 mesh)
Barium carrier, 20 mg $Ba^{+2}$/ml
Cation-exchange resin, Dowex-50W-X8 ($Na^+$ form) (50-100 mesh)
Citrate solution, 3N (pH 6.5)
Diethyl ether
Ethyl alcohol, absolute, 95%
Hydrochloric acid, 12N
Hydrochloric acid-diethyl ether reagent
Nitric acid, 90%, 6N
Sodium carbonate, 3N
Sodium chloride, 4N
Sodium chromate, 3N
Strontium carrier, 20 mg $Sr^{+2}$/ml
Yttrium carrier, 10 mg $Y^{+3}$/ml

*See appendix A.

## Apparatus

Ion-exchange system (fig. 1-1): Connect glass reservoir (graduated, 1-liter separatory funnel) and two glass columns (with extra coarse fritted glass discs in bottom) one above the other, for gravity flow. Fill upper (5-cm diameter) column with 170 ml Dowex-50W resin and lower (1.9-cm diameter) column with 15 ml Dowex-1 resin by pouring into water-filled columns.

Analytical balance
Analytical oven
Desiccator
Millipore filtering apparatus
Millipore #OH membrane-filter (1.5-$\mu$ pore size), 2.5-cm diameter
Separatory funnels, 1 liter, 500 ml
Stainless steel planchets, 2-in. diameter
Whatman #542 filter paper, 2.4-cm diameter
Whatman #4 filter paper, 11-cm diameter

## Procedure

Note: If this analysis follows a strontium-90 determination, begin with step 6 of this procedure.

1. Place 1 liter of milk (note a) into the graduated reservoir. Pipette 1.0 ml each of yttrium, strontium, and barium carriers into 10 ml of citrate solution, swirl to mix and dissolve barium citrate. Transfer quantitatively to milk with 5 ml of distilled water and mix well.

2. Open stopcocks of reservoir, anion column, and cation column, in that order. Control flow rate at 10 ml per minute (check occasionally by collecting effluent for 1 min in graduated cylinder) with anion column stopcock. Stop flow when just enough milk remains in columns to cover resin. Discard effluent milk.

3. Replace milk reservoir with a separatory funnel containing 300 ml of warm distilled water and wash columns with water to displace milk. Stop flow when just enough water remains in columns to cover resin. Discard effluent water.

4. Separate columns: If strontium-90 analysis is desired, process anion column according to procedures from step 5, section I, part A.

5. To the top of cation column, attach a 1-liter separatory funnel containing 1 liter of 4N NaCl. Allow NaCl solution to flow at 10 ml per minute to elute alkali metal and alkaline earth ions and

also to recharge the column. Leave enough eluent to cover resin to a depth of 2 cm. Collect eluate until the total volume is 1 liter. An aliquot of the NaCl eluate can be taken for the determination of stable calcium (sec. I, pt. C). Place 500 ml of distilled water in separatory funnel and let flow into another beaker at 10 ml per minute to remove excess NaCl from the resin (note b). Discard this washing.

6. Dilute remaining NaCl eluate to 1,500 ml with distilled water.

7. Heat solution (85° to 90° C) on a hot plate and add, with stirring, 100 ml of 3N $Na_2CO_3$. Stir gently while on the hot plate to prevent bumping. Remove from hot plate, cool, and decant bulk of supernate. Transfer precipitate to a 250-ml centrifuge bottle. Centrifuge and decant supernate.

8. Wash precipitate twice with 50-ml portions of distilled water. Dry in an oven at 110° C for 4 hours.

9. Remove from oven, cool, and dissolve precipitate in approximately 10 ml of 6N $HNO_3$ (stir vigorously on a magnetic stirrer until dissolved). Filter the solution through Whatman #4 filter paper into a 40-ml graduated centrifuge tube. Discard filter paper and contents.

10. To the filtrate, add a sufficient volume of 90% $HNO_3$ to obtain approximately 70% nitric acid concentration (table 1-1). Stir and cool in an ice bath. Centrifuge and decant supernate.

Table 1-1. Nitric Acid Proportions for Strontium and Barium Precipitations

| To volume of 6N $HNO_3$ | Add amount of 90% $HNO_3$ | To obtain final concentration of |
|---|---|---|
| 8 ml | 15 ml | 69.8% |
| 9 ml | 17 ml | 69.9% |
| 10 ml | 19 ml | 70.0% |
| 11 ml | 21 ml | 70.1% |
| 12 ml | 22 ml | 69.5% |
| 13 ml | 24 ml | 69.6% |
| 14 ml | 26 ml | 69.7% |
| 15 ml | 28 ml | 69.8% |
| 16 ml | 30 ml | 69.8% |

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11. Dissolve precipitate in 5 ml of distilled water. Add 5 ml of ammonium acetate buffer and check for pH of 5 with pH paper. Add additional buffer solution if necessary.

12. To precipitate $BaCrO_4$, heat solution in a water bath and add, dropwise and with stirring, 1 ml of 3N $Na_2CrO_4$. Centrifuge and decant supernate into a 40-ml centrifuge tube. If barium-140 analysis is desired, continue working with precipitate and follow steps 13-18; otherwise, continue working with supernate at step 19.

13. Add approximately 5 ml of 12N HCl to the $BaCrO_4$ precipitate. Heat in a water bath until the yellow $BaCrO_4$ precipitate is dissolved and a white, granular $BaCl_2.2H_2O$ precipitate remains. At this point the solution will have a green coloration from $Cr^{+3}$.

14. Cool in an ice bath and add 20 ml of cold HCl-diethyl ether reagent. Stir, centrifuge, and decant the supernate. A pure white precipitate ($BaCl_2.2H_2O$) should remain. If a coloration occurs, repeat the HCl-diethyl ether application. RECORD THE TIME OF DECANTATION AS START OF LANTHANUM-140 INGROWTH.

15. Weigh a stainless steel planchet and 2.4-cm diameter Whatman #542 filter paper. Place filter paper into filtering apparatus.

16. Transfer the precipitate onto the filter paper with several 10-ml portions of cold absolute ethyl alcohol. Use at least 30 to 40 ml of absolute ethyl alcohol.

17. Transfer filter paper with precipitate to stainless steel planchet, dry at 100° C for 1 hour. Cool in a desiccator 10-15 minutes. Weigh and calculate the recovery of barium carrier as $BaCl_2$ (note c).

18. Count the precipitate on the planchet in a low-background beta counter (note d).

19. To the supernate from step 12, add 30 ml of 90% $HNO_3$, stir, and cool in an ice bath to precipitate strontium nitrate. Centrifuge and decant.

20. Dissolve strontium nitrate precipitate in 10 ml of deionized distilled water. Add 30 ml of 90% $HNO_3$, stir, cool in ice bath, centrifuge, and decant. RECORD THE TIME OF DECANTATION AS START OF YTTRIUM-90 INGROWTH.

21. Dissolve strontium nitrate precipitate in 10 ml of deionized distilled water and add 3 ml of 15N $NH_4OH$. Heat to near boiling in

hot water bath and add 5 ml of 1N ammonium oxalate to precipitate strontium oxalate. Cool in ice bath, centrifuge, and decant.

22. Weigh a stainless steel planchet and 2.5-cm diameter Millipore #OH filter. Place filter into filtering apparatus.

23. Transfer the strontium oxalate precipitate to the filtering apparatus using three 10-ml portions of cold distilled water. Wash precipitate with three 10-ml portions of 95% ethyl alcohol and dry with three 10-ml portions of diethyl ether. Continue suction for 2 minutes.

24. Transfer the filter with precipitate to stainless steel planchet and weigh after a delay of 10-15 minutes. Calculate the recovery of strontium carrier (note e).

25. Count the precipitate for "total radiostrontium" activity.

Notes

a. Milk must be reasonably homogeneous, and should be preserved with formaldehyde and stored under refrigeration (32° to 34° F) for 2 weeks. Large amounts of separated cream or solids will interfere with or prevent flow of milk through resin columns. If clogging occurs, stirring of the resin may increase the flow. For procedure for nonhomogeneous milk, see section I, part D.

b. If resin clogs with extended use, backwashing or slurrying of the resin with distilled water will remove trapped milk particulates.

c. Alternatively, oven drying may be omitted. In such an event, the precipitate is merely left in contact with the atmosphere following the final absolute ethyl alcohol treatment until constant weight is achieved (approximately 3 hours). The recovery of barium carrier is then calculated from the weight of $BaCl_2.2H_2O$.

d. Care must be exercised that filter paper remains centered in planchet to avoid any geometric effects during counting.

e. Alternatively, the flame spectrophotometer may be used to determine recovery of strontium carrier by dissolving the precipitate after it has been counted (sec. I, pt. E).

Calculation

Strontium-89: $pc/l = \frac{1}{(A)(B)} \left[ \frac{(C)}{(D)(E)} - (F)(G)(H) - (G)(I)(J) \right]$

where (A) = counting efficiency in $\frac{cpm}{pc}$ for strontium-89 as strontium oxalate mounted on a 2.5-cm diameter membrane filter

(since 1 pc = 2.22 dpm, $\frac{cpm}{pc} = 2.22 \frac{cpm}{dpm}$),

(B) = correction factor for strontium-89 decay ($e^{-\lambda t}$), where t is the time from sample collection to the time of counting,*

(C) = net cpm of "total radiostrontium",

(D) = recovery of strontium carrier,

(E) = volume of milk sample in liters,

(F) = self-absorption factor for strontium-90 as strontium oxalate mounted on 2.5-cm diameter membrane filter,

(G) = strontium-90 concentration in pc/l,**

(H) = counting efficiency in $\frac{cpm}{pc}$ for strontium-90 as strontium oxalate mounted on 2.5-cm diameter membrane filter,

(I) = counting efficiency in $\frac{cpm}{pc}$ for yttrium-90 as yttrium oxalate mounted on 2.5-cm diameter membrane filter,

(J) = correction factor for yttrium-90 ingrowth ($1-e^{-\lambda t}$), where t is the time from the last decantation of $HNO_3$ from the strontium nitrate precipitate to the time of counting.***

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*Obtained from table 3, "Sr-89 Decay Factors", appendix B.
**Obtained from the calculation in section I, part A.
***Obtained from table 1, "Y-90 Decay and Ingrowth Factors", appendix B.

Barium-140: $$pc/l = \frac{(\text{net cpm})}{[(A)(B)(C)]\ [(D) + (1.15)(E)(F)]}$$

where (A) = recovery of barium carrier,

(B) = volume of milk sample in liters,

(C) = correction factor for decay of barium-140 ($e^{-\lambda t}$), where t is the time from collection of the sample to the time of counting,*

(D) = counting efficiency in $\frac{cpm}{pc}$ for barium-140 on 2.4-cm filter paper

(since 1 pc = 2.22 dpm, $\frac{cpm}{pc} = 2.22 \frac{cpm}{dpm}$),

(E) = counting efficiency in $\frac{cpm}{pc}$ for lanthanum-140 on 2.4-cm filter paper,

(F) = correction factor for the ratio of lanthanum-140 activity to barium-140 activity ($1-e^{-0.015t}$), where t is the time in hours from the barium separation to the time of counting.** The factor 1.15 in the denominator of the above equation is the ratio of lanthanum-140 activity to barium-140 activity in equilibrium.

## C. Calcium

### Principle of Method

Milk is passed through cation- and anion-exchange resin columns. Strontium, barium, and calcium are adsorbed on the cation-exchange resin, then eluted with sodium chloride solution. An aliquot of this solution is then diluted to reduce the high sodium ion concentration. From this diluted aliquot, calcium oxalate is precipitated, dissolved in dilute hydrochloric acid, and titrated against standardized potassium permanganate.

### Reagents***

Ammonium hydroxide, 6N
Ammonium oxalate, 0.03N

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*Obtained from table 4, "Ba-140 Decay Factors", appendix B.
**Obtained from table 5, "La-140 Ingrowth Factors", appendix B.
***See appendix A.

Anion-exchange resin, Dowex-1-X8 ($Cl^-$ form) (50-100 mesh)
Barium carrier, 20 mg $Ba^{+2}$/ml
Cation-exchange resin, Dowex-50W-X8 ($Na^+$ form) (50-100 mesh)
Citrate solution, 3N (pH 6.5)
Hydrochloric acid, 6N
Oxalic acid, 1N
Potassium permanganate, 0.05N (note a)
Sodium chloride, 4N
Sodium oxalate standard
Strontium carrier, 20 mg $Sr^{+2}$/ml
Yttrium carrier, 10 mg $Y^{+3}$/ml

## Apparatus

Ion-exchange system (fig. 1-1): Connect glass reservoir (separatory funnel) and two glass columns (with extra coarse fritted glass discs in bottom) one above the other, for gravity flow. Fill upper (5-cm diameter) column with 170 ml Dowex-50W resin, and lower (1.9-cm diameter) column with 15 ml Dowex-1 resin, by pouring resin into water-filled columns.
Analytical balance
Burettes, 50 ml
Magnetic stirrer hot plate (stirring bars)

## Procedure

1. Follow the procedure given in section I, part A, steps 1-4a.

2. Pipette a 20-ml aliquot of the initial eluate collected in step 4a, section I, part A, into a 40-ml glass centrifuge tube.

3. Dilute the 20-ml aliquot to approximately 30 ml with distilled water.

4. Heat in a hot water bath and add 10 ml of 1N oxalic acid and stir. Adjust to pH 3 while hot with 6N $NH_4OH$ (use pH meter) to precipitate calcium oxalate. Cool slowly to room temperature, centrifuge, and discard the supernate.

5. Wash the precipitate and the wall of the centrifuge tube thoroughly, using not more than 10 ml of 0.03N ammonium oxalate. Centrifuge and discard the supernate.

6. Wash the precipitate with 10 ml of hot distilled water. Cool slowly to room temperature, centrifuge, and discard supernate (note b).

7. Dissolve the precipitate in approximately 2.5 ml of 6N HCl with gentle heating.

8. Dilute to a volume of approximately 10 ml with distilled water. Transfer quantitatively to a 125-ml Erlenmeyer flask, rinsing the centrifuge tube with distilled water.

9. Add an additional 1 ml of 6N HCl and adjust the volume of solution to approximately 25 ml. Heat to just below boiling.

10. Titrate while hot with standardized 0.05N $KMnO_4$ to the first faint pink end-point which persists for at least 30 seconds.

Notes

a. Standardize against NBS sodium oxalate standard. Weigh out in triplicate 80 mg of dry sodium oxalate standard. Dissolve each in 25 ml of distilled water containing 2 ml of 6N HCl. Titrate each with the approximately 0.05N $KMnO_4$ to a faint pink end-point which persists for at least 30 seconds. Standardize $KMnO_4$ at least once a week.

$$\underline{N}\ KMnO_4 = \frac{g\ \text{sodium oxalate standard}}{(\text{ml of } KMnO_4)\ (0.067)}$$

b. It is important that all excess oxalic acid be removed from the precipitate. A stirring rod may be used to agitate the precipitate.

Calculation

$$g\ Ca/l\ \text{of milk} = \frac{(\text{ml of } KMnO_4)\ \overline{(\underline{N}\ KMnO_4)}\ (\text{meq wt Ca})}{(\text{aliquot vol in ml})}$$

where $(\text{ml of } KMnO_4)$ = volume of $KMnO_4$ solution used for titration,

$\overline{(\underline{N}\ KMnO_4)}$ = average normality of $KMnO_4$ solution (average of three values from standardization),

(meq wt Ca) = milli-equivalent weight of calcium.

For the particular system described in the procedure, the equation reduces to

$$g\ Ca/l\ \text{of milk} = (\text{ml of } KMnO_4)\ \overline{(\underline{N}\ KMnO_4)}$$

## D. Strontium-90, Strontium-89, and Barium-140 in Nonhomogeneous Milk

This method may be used for milk which is no longer homogeneous, i.e., soured, curdled, or containing excessive cream.

### Principle of Method

Milk, with added strontium and barium carriers, is thoroughly mixed with cation-exchange resin. The resin is washed and placed in a column. The alkaline earths are eluted with sodium chloride solution, precipitated as the carbonates, and converted to the nitrates, precipitating strontium and barium nitrate. The nitrate precipitate is dissolved and barium is precipitated as the chromate, purified as the chloride, then counted to determine barium-140. Strontium is precipitated from the supernate as the nitrate, dissolved in deionized distilled water, reprecipitated as the nitrate, and stored for yttrium-90 ingrowth. The strontium nitrate is dissolved, yttrium carrier added, and strontium reprecipitated from concentrated nitric acid. Yttrium is extracted from the supernate with tributyl phosphate, reextracted with dilute nitric acid, and precipitated as the oxalate. The yttrium oxalate is weighed to determine recovery of yttrium carrier, then counted. The strontium nitrate is dissolved and precipitated as strontium oxalate. The oxalate precipitate is filtered, weighed to determine strontium carrier recovery, and counted for "total radiostrontium". The strontium-89 activity is calculated from the difference between the count for "total radiostrontium" and the count due to the previously determined strontium-90 activity.

### Reagents*

Ammonium acetate buffer, pH 5
Ammonium hydroxide, 15N
Ammonium oxalate, 1N
Barium carrier, 20 mg $Ba^{+2}$/ml
Cation-exchange resin, Dowex-50W-X8 ($Na^{+}$ form) (50-100 mesh)
Diethyl ether
Ethyl alcohol, absolute, 95%
Hydrochloric acid, 12N
Hydrochloric acid-diethyl ether reagent
Nitric acid, 90%, 16N, 14N, 8N, 6N, 0.1N
Oxalic acid, 2N
Sodium carbonate, 3N
Sodium chloride, 4N

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*See appendix A.

Sodium chromate, 3N
Strontium carrier, 20 mg $Sr^{+2}$/ml
Tributyl phosphate (TBP), preequilibrated
Yttrium carrier, 10 mg $Y^{+3}$/ml

## Apparatus

Analytical balance
Analytical oven
Desiccator
Glass column, with fritted glass bottom, 5-cm diameter
Infrared lamp
Magnetic stirrer (stirring bars)
Millipore filtering apparatus
Millipore #OH membrane-filter (1.5-$\mu$ pore size), 2.5-cm diameter
pH meter
Separatory funnels, 500 ml, 60 ml
Stainless steel planchets, 2-in. diameter
Whatman #542 filter paper, 2.4-cm diameter
Whatman #4 filter paper, 11-cm diameter

## Procedure

1. Pipette 1.0 ml each of strontium and barium carriers to 1 liter of nonhomogeneous milk in a 2-liter beaker containing 140 ml of cation-exchange resin ($Na^+$ form).

2. Stir for 1 hour with magnetic stirrer. Let resin settle and decant supernate. Wash milk from the resin with warm distilled water and decant the water (note a).

3. Transfer resin to the glass column. To the top of the column attach a 500-ml separatory funnel containing 500 ml of 4N NaCl.

4. Allow NaCl eluent to flow at 10 ml per minute to wash and recharge the resin. When only enough eluent remains to cover resin to a depth of 2 cm, place 500 ml of distilled water in the separatory funnel and allow to flow at 10 ml per minute until combined NaCl-water eluate has a volume of 1 liter.

5. Dilute eluate to 1,500 ml with distilled water. Heat the solution (85° to 90° C) on a hot plate and add, with stirring, 100 ml of 3N $Na_2CO_3$. Stir gently while on the hot plate to prevent bumping. Remove from hot plate, cool, and decant bulk of supernate. Transfer precipitate to a 250-ml centrifuge bottle. Centrifuge and decant supernate.

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6. Wash precipitate twice with 50-ml portions of distilled water. Dry in oven at 110° C for a minimum of 4 hours.

7. Remove from oven, cool, and dissolve precipitate in approximately 10 ml of 6N $HNO_3$ (stir vigorously with a magnetic stirrer until dissolved). Filter the solution through Whatman #4 filter paper into a 40-ml graduated centrifuge tube. Discard filter paper and contents.

8. To the filtrate, add a sufficient volume of 90% $HNO_3$ to obtain a 70% nitric acid concentration (table 1-1). Stir and cool in an ice bath. Centrifuge and decant supernate.

9. Dissolve precipitate in 5 ml of distilled water. Add 5 ml of ammonium acetate buffer and check for pH of 5 with pH paper. Add additional buffer solution if necessary.

10. To precipitate $BaCrO_4$, heat solution in a water bath and add, dropwise with stirring, 1 ml of 3N $Na_2CrO_4$. Centrifuge and decant supernate into a 40-ml centrifuge tube. If barium-140 analysis is desired, continue working with precipitate and follow steps 11-16; otherwise, continue working with supernate at step 17.

11. Add approximately 5 ml of 12N HCl to the $BaCrO_4$ precipitate. Heat in a water bath until the yellow $BaCrO_4$ precipitate is dissolved and a white, granular $BaCl_2.2H_2O$ precipitate remains. At this point, the solution will have a green coloration from $Cr^{+3}$.

12. Cool in an ice bath and add 20 ml of cold HCl-diethyl ether reagent. Stir, centrifuge, and decant the supernate. A pure white precipitate ($BaCl_2.2H_2O$) should remain. If a coloration occurs, repeat the HCl-diethyl ether application. RECORD THE TIME OF DECANTATION AS THE START OF LANTHANUM-140 INGROWTH.

13. Weigh a stainless steel planchet and 2.4-cm diameter Whatman #542 filter paper. Place filter paper into Millipore filtering apparatus.

14. Transfer the precipitate onto the filter paper with several 10-ml portions of cold absolute ethyl alcohol. Use at least 30 to 40 ml of absolute ethyl alcohol.

15. Transfer filter paper with precipitate to stainless steel planchet, dry at 100° C for 1 hour. Cool in a desiccator 10-15 minutes. Weigh and calculate the recovery of barium carrier as $BaCl_2$ (note b).

16. Count the precipitate on the planchet in a low-background beta counter (note c).

17. To the supernate from step 10, add 30 ml of 90% $HNO_3$, stir, and cool in ice bath. Centrifuge and decant. Discard supernate.

18. Dissolve the strontium nitrate precipitate in 10 ml of deionized distilled water. Add 30 ml of 90% $HNO_3$, stir, cool in ice bath, centrifuge, and decant. RECORD THE TIME OF DECANTATION AS THE START OF YTTRIUM-90 INGROWTH. Drain excess $HNO_3$ from the centrifuge tube by inverting on a paper towel.

19. Store strontium nitrate precipitate for ingrowth of yttrium-90 (usually 1-2 weeks).

20. After the ingrowth period, pipette 1.0 ml of yttrium carrier into the centrifuge tube and add 1.0 ml of 8N $HNO_3$. Warm gently to dissolve precipitate. Add 10 ml 90% $HNO_3$ and cool in ice bath for 10 minutes. Centrifuge and quantitatively decant the supernate into a 60-ml separatory funnel. RECORD THE TIME OF SEPARATION AS START OF YTTRIUM-90 DECAY IN THE SUPERNATE AND YTTRIUM-90 INGROWTH IN THE TOTAL STRONTIUM PRECIPITATE. Save the precipitate. Drain excess $HNO_3$ from centrifuge tube by inverting tube on a paper towel.

21. To the solution in the separatory funnel, add 4 ml of distilled water to make the solution 14N in nitric acid. Add 10 ml of preequilibrated TBP to this solution.

22. Equilibrate by shaking for approximately 2-3 minutes; allow phases to separate. Drain acid phase away and discard.

23. Add 15 ml of 14N $HNO_3$ to the separatory funnel and equilibrate again for approximately 2-3 minutes; allow phases to separate. Drain acid phase away and discard.

24. Repeat step 23.

25. Remove yttrium from the organic phase by adding 15 ml of distilled water and equilibrating for approximately 2-3 minutes. Allow phases to separate and drain aqueous phase into a 40-ml centrifuge tube.

26. Repeat step 25 using 15 ml of 0.1N $HNO_3$ instead of distilled water and drain acid phase into same 40-ml centrifuge tube as in step 25.

27. To precipitate yttrium oxalate, add 5 ml of 2N oxalic acid and adjust to pH 1.5 with approximately 2 ml of 15N $NH_4OH$ (using pH meter). Stir and cool in an ice bath approximately 20 minutes. Centrifuge and discard supernate.

28. Weigh planchet and 2.5-cm diameter membrane filter. Place membrane filter into filtering apparatus.

29. Take up precipitate in 10 ml of cold water and transfer to filtering apparatus. Wash with one 10-ml portion of cold distilled water using the water to wash out centrifuge tube. Dry precipitate with three 20-ml portions of 95% ethyl alcohol and three 10-ml portions of diethyl ether. Continue suction for 2 minutes.

30. Transfer membrane filter with precipitate to stainless steel planchet and weigh after a delay of 10-15 minutes. Calculate the recovery of yttrium carrier.

31. Count the yttrium oxalate precipitate in a low-background beta counter (note c). Count again after 3 days to confirm absence of contamination of yttrium-90 by other radionuclides.

32. Dissolve strontium nitrate precipitate (obtained in step 20) in 10 ml of deionized distilled water and add 3 ml of 15N $NH_4OH$. Heat to near boiling in hot water bath and add 5 ml of 1N ammonium oxalate to precipitate strontium oxalate. Cool in ice bath, centrifuge, and decant.

33. Weigh a stainless steel planchet and 2.5-cm diameter Millipore #OH filter. Place filter into filtering apparatus.

34. Transfer the strontium oxalate precipitate to the filtering apparatus using three 10-ml portions of cold distilled water. Wash precipitate with three 10-ml portions of 95% ethyl alcohol and dry with three 10-ml portions of diethyl ether. Continue suction for 2 minutes.

35. Transfer the filter with precipitate to stainless steel planchet and weigh after a delay of 10-15 minutes. Calculate the recovery of strontium carrier (note d).

36. Count the precipitate for "total radiostrontium" activity.

Notes

a. Calcium determination should not be made by this method unless a quantitative recovery of the resin can be made.

b. Alternatively, oven drying may be omitted. In such an event, the precipitate is merely left in contact with the atmosphere, following the final absolute alcohol treatment, until constant weight is achieved. The recovery of barium carrier is then calculated from the weight of $BaCl_2.2H_2O$.

c. Care must be exercised that filter paper or membrane filter remains centered in planchet to avoid any geometric effects during counting.

d. Alternatively, the flame spectrophotometer may be used to determine recovery of strontium carrier by dissolving the precipitate after it has been counted. See section I, part E.

Calculation

Strontium-90:

$$pc/l = \frac{(\text{net cpm})}{(A)(B)(C)(D)(E)(F)}$$

where (A) = recovery of strontium carrier,

(B) = recovery of yttrium carrier,

(C) = counting efficiency in $\frac{cpm}{pc}$ for yttrium-90 as yttrium oxalate mounted on a 2.5-cm membrane filter

(since 1 pc = 2.22 dpm, $\frac{cpm}{pc} = 2.22 \frac{cpm}{dpm}$),

(D) = sample volume in liters,

(E) = correction factor for yttrium-90 ingrowth $(1-e^{-\lambda t})$, where t is the time from decantation of $HNO_3$ from strontium $(NO_3)_2$ precipitate to the time of decantation of yttrium-90 supernate,*

(F) = correction factor for yttrium-90 decay $(e^{-\lambda t})$, where t is the time from the decantation of yttrium-90 supernate to the time of counting.**

Strontium-89:

Same calculation as in section I, part B.

Barium-140:

Same calculation as in section I, part B.

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*Obtained from table 2, "Y-90 Ingrowth Factors", appendix B.
**Obtained from table 1, "Y-90 Decay and Ingrowth Factors", appendix B.

## E. Flame Photometric Determination of the Recovery of Strontium Carrier

### Principle of Method

Strontium oxalate, precipitated in other procedures, is dissolved in deionized distilled water. Strontium is determined flame photometrically at a wavelength of 460 mμ using an oxyacetylene flame.

### Reagents*

$HNO_3$, 6N
Strontium standard solution, 0.4 mg $Sr^{+2}$/ml

### Apparatus

Beckman DU spectrophotometer, line-operated and equipped with flame attachment, spectral energy recording attachment, photomultiplier, and Sargent SR recorder
Atomizer burners
Beakers, 5 ml

### Procedure

Note: Approximate settings for instruments are:

| | |
|---|---|
| Wavelength | 460 mμ |
| Slit width | 0.04-0.08 mm |
| Sensitivity (power supply) | 7-9 |
| Oxygen pressure | 3 lb* |
| Acetylene pressure | 10 lb** |
| Selector switch | 1 |
| Load resistor | 2 |
| Variable range (recorder) | 40-100 mv |

*Oxygen pressure is given on burner.
**Adjusted so that solution is atomized at 1.5 to 2 ml per minute.

*See appendix A.

1. Dissolve strontium oxalate precipitate (on membrane filter) in 1 ml of 6N $HNO_3$. Transfer solution quantitatively to 25-ml volumetric flask with deionized distilled water and dilute to volume.

2. Transfer a 4-ml aliquot of the strontium standard solution into a 5-ml beaker.

3. Atomize the solution into the flame and adjust the recorder response so that recorder reading is at approximately half scale.

4. Transfer 4 ml of deionized distilled water to a 5-ml beaker.

5. Atomize and record the response.

6. Transfer a 4-ml aliquot of the sample solution into a 5-ml beaker.

7. Atomize the solution into the flame and record the response.

Calculation

$$\text{Strontium carrier recovery} = \frac{(0.5)\,(A-B)}{(C-B)}$$

where (A) = sample reading,

(B) = baseline reading from deionized water blank,

(C) = standard reading.

The factor 0.5 in the equation results from the dilution of the recovered strontium by a factor of 25 and the dilution of the carrier strontium solution by a factor of 50 in making the standard strontium solution.

## F. Flame Photometric Determination of Potassium

Principle of Method

Milk containing acetic acid is heated in a water bath until the casein separates. The solution is centrifuged, filtered, and passed through a cation-exchange resin ($H^+$ form) column. Potassium, sodium, calcium, and magnesium are quantitatively adsorbed on the resin. These elements are eluted from the resin with

hydrochloric acid and the concentration of potassium in the eluate is determined by flame photometry (note a).

Reagents*

Acetic acid, glacial
Cation-exchange resin, Dowex-50W-X8 ($H^+$ form) (50-100 mesh)
Hydrochloric acid, 3.5N, 0.5N
Potassium blank solution
Potassium standard solution

Apparatus

Beckman DU spectrophotometer, line operated and equipped with flame attachment, spectral energy recording attachment, photomultiplier and Sargent SR recorder
Atomizer burners
Beakers, 5 ml
Glass column, with fritted glass bottom, 1.9-cm diameter
Whatman #541 filter paper, 11-cm diameter

Procedure

1. Fill 1.9-cm diameter glass column with 10 ml Dowex-50W-X8 ($H^+$ form) by pouring resin into water-filled column.

2. Measure 60 ml of milk in graduated cylinder. Wash milk from cylinder into a graduated 250-ml beaker using deionized distilled water and dilute to approximately 100 ml.

3. Add 2 ml of glacial acetic acid and stir. Heat in water bath at 80° C for 10 minutes or until casein separates from the milk serum.

4. Dilute to 150 ml with deionized distilled water. Transfer to a 250-ml centrifuge bottle (do not wash out the beaker). Centrifuge.

5. Decant supernate into a beaker and filter through Whatman #541 filter paper. Collect 100 ml of this solution in a graduated cylinder (note b).

6. Transfer the 100-ml sample quantitatively to a separatory funnel using deionized distilled water.

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*See appendix A.

7. Attach the separatory funnel to the glass column filled with cation resin. Let solution pass over the resin at a flow rate of 4 ml per minute. Discard effluent.

8. Wash the resin with 30 ml of deionized distilled water, discarding the wash water.

9. Attach a separatory funnel containing 75 ml of 3.5N HCl to the top of the column. Let acid flow at 2 ml per minute. Discard the first 3 ml of effluent. Collect the next 50 ml of eluate in a volumetric flask.

10. Allow the remaining 3.5N HCl to flow through to recharge the resin. Discard effluent. Wash resin with 50 ml of deionized distilled water and discard effluent water.

11. Pipette 3 ml of the 50-ml eluate into a 100-ml volumetric flask. Dilute to volume with 0.5N HCl.

12. Use this 3:100 dilution to determine the concentration of potassium.

Preparation of Calibration Curve

1. Pipette 3.0, 4.0, 5.0, 6.0, 7.0, and 8.0 ml, respectively, of the potassium standard solution into each of six 200-ml volumetric flasks. These represent 15, 20, 25, 30, 35, and 40 ppm potassium.

2. Pipette 2 ml of the potassium blank solution into each volumetric flask.

3. Dilute to volume with 0.5N HCl and mix thoroughly.

4. Fill a 5-ml beaker with deionized distilled water and atomize. Record the response to determine the background reading (note c).

5. Fill 5-ml beakers with each of the potassium standard solutions. Atomize each and record the response. Atomize a background sample of deionized distilled water between each standard solution.

6. Subtract the background response readings from the standard response readings. Plot on linear graph paper with potassium ion concentration (ppm) as the abscissa and relative response reading as the ordinate.

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Flame Determination of Sample

1. Fill a 5-ml beaker with deionized distilled water and atomize. Record the background response.

2. Fill a 5-ml beaker with the sample solution. Atomize and record the response. Subtract the background response from the sample response reading and compare to the calibration curve.

3. Read the ppm of potassium contained in the sample from the calibration curve. Multiply ppm by 0.0416 g/l-ppm to obtain g of potassium per liter of milk. This conversion factor takes into account the dilutions in the procedure and the relationship of ppm to g/l.

Notes

a. With the appropriate standards for the flame photometer calibration curve, this method may also be used for sodium, calcium, and magnesium determinations.

b. Filtering the sample is necessary to remove small particles of the casein which may not settle out when centrifuged.

c. Approximate settings for the instruments used are:

| | |
|---|---|
| Wavelength | 766 m$\mu$ |
| Slit width | 3.0-3.5 mm |
| Sensitivity (power supply) | 9 |
| Oxygen pressure | 3 lb* |
| Acetylene pressure | 10 lb** |
| Selector switch | 1 |
| Load resistor | 2 |
| Variable range (recorder) | 40-100 mv |

*Oxygen pressure is given on burner.
**Adjusted so that solution is atomized at 1.5 to 2 ml per minute.

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## SECTION II. FOOD, BIOTA, SILT, AND SOIL

### A. Gross Alpha and Beta Activity

Principle of Method

The sample is dried and converted to ash in a muffle furnace. The ash is packed in a planchet and counted to determine alpha and beta activity.

Apparatus

Analytical balance
Analytical oven
Desiccator
Dishes, Pyrex or Corningware
Electric blender
Muffle furnace
Pan balance
Stainless steel planchets, 2-in. diameter

Procedure

Note: For required sample size, see table 2-1.
For samples other than food, omit step 1.

1. Weigh food sample. Check for and remove inedible objects. Transfer solid food to a 10-gallon stainless steel container and blend for 20 minutes in an electric blender. Add liquid portions (except milk) and blend for 10 minutes.

2. Transfer sample into preweighed Pyrex dishes (Corningware for food). When sample contains preservatives (alcohol, formaldehyde, etc.), the preservative is decanted away and saved. After weighing sample, the preservative is added back to sample and entire mixture is dried.

3. Dry in oven at 100° to 120° C for 48 hours.

4. Transfer to muffle furnace and raise temperature slowly to 520° C. Ash at 520° C for 96 hours.

Table 2-1. Required Amount and Approximate Ash Content of Various Sample Media

| Media | Required amount* | Approximate ash content |
|---|---|---|
| Apples | 13.0 kg | 0.3% |
| Bananas | 4.0 kg | 0.9% |
| Beans (green, snap) | 5.5 kg | 0.6% |
| (kidney, dry) | 2.5 kg | 1.6% |
| (large lima, dry) | 2.5 kg | 1.6% |
| (pinto, dry) | 2.5 kg | 1.8% |
| Beef (meat) | 5.0 kg | 0.7% |
| Bones | 0.2 kg | 18.0% |
| Bread | 1.7 kg | 2.0% |
| Cabbage | 7.0 kg | 0.5% |
| Carrots | 3.5 kg | 1.1% |
| Celery | 4.5 kg | 0.8% |
| Cereals (All Bran) | 0.5 kg | 8.2% |
| (Cheerios) | 1.0 kg | 4.5% |
| (Corn Flakes) | 1.5 kg | 3.1% |
| (Grape-Nuts) | 1.5 kg | 2.6% |
| (Rice Krispies) | 1.5 kg | 3.1% |
| (Shredded Wheat) | 2.5 Kg | 1.4% |
| (Special "K") | 1.5 kg | 3.0% |
| (Wheaties) | 1.0 kg | 4.8% |
| Coffee (instant) | 0.4 kg | 12.0% |
| (regular, brewed) | 1.8 kg | 0.2% |
| Crab (whole) | 0.5 kg | 14.5% |
| Diet Foods (composite) | 3.5 kg | 1.0% |
| Eggplant | 5.0 kg | 0.8% |
| Eggs | 5.0 kg | 0.7% |
| Fish (whole) | 0.5 kg | 6.5% |
| (edible, red snapper) | 2.0 kg | 1.8% |
| Grass (dry) | 1.0 kg | 5.5% |
| (green) | 2.0 kg | 2.0% |
| Lettuce | 7.0 kg | 0.5% |
| Milk (evaporated) | 2.5 kg | 1.6% |
| (fresh) | 5.0 kg | 0.7% |
| Oleomargarine | 1.6 kg | 2.3% |
| Oranges | 9.0 kg | 0.4% |
| Oysters (meat) | 0.5 kg | 6.0% |
| Pork (meat) | 5.0 kg | 0.8% |
| Potatoes (fresh) | 4.0 kg | 1.0% |
| Pumpkins | 8.5 kg | 0.4% |
| Rice (white) | 7.0 kg | 0.5% |
| Rutabagas | 6.0 kg | 0.6% |
| Shrimp (whole) | 1.0 kg | 4.5% |
| Silt | 1.0 kg | variable |
| Squash | 5.0 kg | 0.7% |
| Tomatoes | 4.5 kg | 0.8% |
| Watermelon | 13.0 kg | 0.3% |

*In order to have sufficient ash for the chemical procedures listed in this manual.

5. Cool, weigh, and grind ash to a fine powder.

6. Transfer a small aliquot of the ash to a weighed stainless steel planchet. Distribute the ash evenly and pack down. Use as little ash as possible, but use sufficient ash to allow even distribution and firm packing. Dry in oven at 110° C for 2-3 hours, and cool in a desiccator for 1 hour (until constant weight). Weigh sample and planchet. Store sample in desiccator (silica gel desiccant) until sample is to be counted.

7. Count to determine alpha and beta activity.

Calculation

Gross Beta:

$$\text{pc/g ash} = \frac{\text{(net beta cpm)}}{\text{(A)(B)(C)}}$$

where (A) = counting efficiency in $\frac{\text{cpm}}{\text{pc}}$ for gross beta activity (calibrated with equilibrium mixture of strontium-90/yttrium-90)

(since 1 pc = 2.22 dpm, $\frac{\text{cpm}}{\text{pc}} = 2.22 \frac{\text{cpm}}{\text{dpm}}$),

(B) = weight of ash in planchet in grams,

(C) = correction factor for self-absorption in ash sample.

Gross Alpha:

$$\text{pc/g ash} = \frac{\text{(net alpha cpm)}}{\text{(A) (B) (C)}}$$

where (A) = alpha counting efficiency in $\frac{\text{cpm}}{\text{pc}}$ for a "weightless" sample

(since 1 pc = 2.22 dpm, $\frac{\text{cpm}}{\text{pc}} = 2.22 \frac{\text{cpm}}{\text{dpm}}$),

(B) = weight of ash in planchet in grams,

(C) = correction factor for self-absorption in ash sample.

## B. Strontium-90, Strontium-89, and Barium-140

### Principle of Method

An aliquot of an ashed sample, to which barium and strontium carriers have been added, is fused with sodium hydroxide and sodium carbonate. The fusion mix is taken up in distilled water, leaving the alkaline earth carbonates in the precipitate. The precipitate is dissolved in dilute hydrochloric acid. The alkaline earths are reprecipitated as the carbonates, then converted to the nitrates, precipitating strontium and barium nitrate. The nitrate precipitate is dissolved in distilled water and the solution scavenged by precipitation of mixed hydroxides with dilute ammonium hydroxide. Barium is precipitated as the chromate, purified as the chloride, then counted to determine barium-140. Strontium is precipitated from the supernate as the nitrate and stored for yttrium-90 ingrowth. The strontium nitrate is dissolved, yttrium carrier added, and strontium reprecipitated from concentrated nitric acid. Yttrium is extracted from the supernate with tributyl phosphate, reextracted with dilute nitric acid, and precipitated as the oxalate. The yttrium oxalate is weighed to determine recovery of yttrium carrier, then counted. The strontium nitrate is dissolved in deionized distilled water and reprecipitated as the nitrate. The nitrate is dissolved and precipitated as strontium oxalate. The oxalate precipitate is filtered, weighed to determine strontium carrier recovery, and counted for "total radiostrontium". The strontium-89 activity is calculated from the difference between the count for "total radiostrontium" and the count due to the previously determined strontium-90 activity.

### Reagents*

Ammonium acetate buffer, pH 5
Ammonium hydroxide, 15N, 6N
Ammonium oxalate, 1N
Barium carrier, 20 mg $Ba^{+2}$/ml
Diethyl ether
Ethyl alcohol, absolute, 95%
Hydrochloric acid, 12N, 6N
Hydrochloric acid-diethyl ether reagent
Mixed rare earth carrier
Nitric acid, 90%, 14N, 8N, 6N, 0.1N
Oxalic acid, 2N
Sodium carbonate, anhydrous
Sodium carbonate, 3N, 0.1N
Sodium chromate, 3N

*See Appendix A.

Sodium hydroxide, pellets
Strontium carrier, 20 mg $Sr^{+2}$/ml
Tributyl phosphate (TBP), preequilibrated
Yttrium carrier, 10 mg $Y^{+3}$/ml

## Apparatus

Analytical balance
Analytical oven
Desiccator
Dishes, Pyrex or Corningware
Electric blender
Millipore filtering apparatus
Millipore #OH membrane filter (1.5-$\mu$ pore size), 2.5-cm diameter
Muffle furnace
pH meter
Separatory funnels, 60 ml
Stainless steel planchets, 2-in. diameter
Whatman #541 filter paper, 11-cm diameter
Whatman #542 filter paper, 2.4-cm diameter

## Procedure

1. Weigh an aliquot (1-10 g, depending on the silica content) of ash into a 250-ml nickel crucible (steps 1-5, sec. II, pt. A). RECORD WEIGHT OF ASH. Pipette in 1.0 ml of strontium carrier and 1.0 ml of barium carrier. Mix thoroughly with a nickel spatula. Add to this mixture 50 g of NaOH pellets (5 g for each g of ash) and mix thoroughly. Fuse over a Meeker burner for 20-30 minutes and then slowly stir in 5 g of anhydrous $Na_2CO_3$ (0.5 g for each g of ash). Continue heating the clear red melt for 20-30 minutes.

2. Remove the crucible from flame with long-handled tongs and cool in a cold water bath. CAUTION: Take up the cooled fusion-mixture in about 200 ml of hot distilled water. Disintegrate by gentle boiling. Transfer the mixture to a 250-ml centrifuge bottle with distilled water. Centrifuge 5 minutes and discard the supernate. Add 40 ml of 3N $Na_2CO_3$. Heat 10 minutes. Centrifuge and discard the supernate. Repeat $Na_2CO_3$ wash.

3. Dissolve the precipitate in a minimal amount of 6N HCl. Heat to boiling on a hot plate to expel $CO_2$ and aid dissolution. Make the solution pH 3 with 6N $NH_4OH$. Centrifuge and decant the supernate into a 40-ml centrifuge tube.

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4. Dissolve the precipitate in a minimal amount of 6N HCl. Warm gently for a few seconds. Reprecipitate with 6N $NH_4OH$ (pH 3). Centrifuge and combine the supernate with that from step 3. If the combined supernates are too large, use a second 40-ml centrifuge tube.

5. Repeat step 4. Discard the precipitate.

6. To precipitate the alkaline earths, add 10 to 15 ml of 3N $Na_2CO_3$ to the supernate in each tube obtained from steps 3, 4, and 5. Digest the precipitates in a water bath for about 30 minutes. Centrifuge and check supernates for completeness of precipitation with a few drops of 3N $Na_2CO_3$. Discard the supernates. Dry the precipitates in an oven at 110° C for 3 hours or until completely dry.

7. Remove from oven, cool, and dissolve each of the precipitates in approximately 10 ml of 6N $HNO_3$ (warming in a water bath with occasional stirring may aid dissolution). To the solution, add a sufficient volume of 90% $HNO_3$ to obtain a 70% nitric acid concentration (table 1-1). Cool in ice bath 5 minutes, centrifuge, and discard the supernate.

8. Dissolve the combined precipitate in about 20 ml of distilled water, and add 1 ml of mixed rare earth carrier; adjust to pH 7 with 6N $NH_4OH$. Heat, stir, and filter through Whatman #541 filter paper. Discard the mixed hydroxide precipitate.

9. Add 5 ml of ammonium acetate buffer to the filtrate from step 8. Check for pH of 5 with pH paper. Add additional buffer solution if necessary.

10. To precipitate $BaCrO_4$, heat solution in a water bath and add, dropwise with stirring, 1 ml of 3N $Na_2CrO_4$. Centrifuge and decant supernate into a 40-ml centrifuge tube. If barium-140 analysis is desired, continue working with precipitate and follow steps 11-16; otherwise, continue working with supernate at step 17.

11. Add approximately 5 ml of 12N HCl to the $BaCrO_4$ precipitate. Heat in a water bath until the yellow $BaCrO_4$ precipitate is dissolved and a white, granular $BaCl_2.2H_2O$ precipitate remains. At this point, the solution will have a green coloration from $Cr^{+3}$.

12. Cool in an ice bath and add 20 ml of cold HCl-diethyl ether reagent. Stir, centrifuge, and decant the supernate. A pure white precipitate ($BaCl_2.2H_2O$) should remain. If a coloration occurs,

repeat the HCl-diethyl ether application. RECORD THE TIME OF DECANTATION AS THE START OF LANTHANUM-140 INGROWTH.

13. Weigh a stainless steel planchet and 2.4-cm diameter Whatman #542 filter paper. Place filter paper into filtering apparatus.

14. Transfer the precipitate onto the filter paper with several 10-ml portions of cold absolute ethyl alcohol. Use at least 30 to 40 ml of absolute ethyl alcohol.

15. Transfer filter paper with precipitate to stainless steel planchet. Leave the precipitate in contact with the atmosphere until constant weight is achieved; usually 2 hours is sufficient time. Weigh and calculate the recovery of barium carrier as $BaCl_2.2H_2O$ (note a).

16. Count the precipitate in the planchet for barium-140 (note b).

17. Adjust supernate from step 10 to pH 7 with 6N $NH_4OH$. Check with pH paper. Add sufficient 3N $Na_2CO_3$ (5 ml) to the supernate to precipitate strontium carbonate. Centrifuge and check supernate for completeness of precipitation with a few drops of $Na_2CO_3$. Decant the supernate. Wash the strontium carbonate precipitate with 0.1N $Na_2CO_3$. Centrifuge and decant the supernate.

18. Dissolve carbonate precipitate in 5 ml of 6N $HNO_3$. Add 20 ml of 90% $HNO_3$, cool in ice bath for 10 minutes, centrifuge, and decant supernate.

19. Dissolve the strontium nitrate precipitate in 10 ml of deionized distilled water and add 30 ml of 90% $HNO_3$. Cool in ice bath, centrifuge, and decant. RECORD THE TIME OF DECANTATION AS START OF YTTRIUM-90 INGROWTH. Drain excess acid on paper towels.

20. Store strontium nitrate precipitate for ingrowth of yttrium-90 (usually 1-2 weeks).

21. After the ingrowth period, pipette 1.0 ml of yttrium carrier into the centrifuge tube and add 1.0 ml of 8N $HNO_3$. Warm gently to dissolve precipitate. Add 10 ml of 90% $HNO_3$ and cool in ice bath for 10 minutes. Centrifuge and quantitatively decant the supernate into a 60-ml separatory funnel. RECORD THE TIME OF SEPARATION AS START OF YTTRIUM-90 DECAY AND START OF YTTRIUM-90 INGROWTH IN THE TOTAL STRONTIUM PRECIPITATE. Save the precipitate. Drain excess $HNO_3$ from centrifuge tube by inverting tube on paper towels.

22. To the solution in the separatory funnel, add 4 ml of distilled water to make the solution 14N in nitric acid. Add 10 ml of preequilibrated TBP to this solution.

23. Equilibrate by shaking for approximately 2-3 minutes; allow phases to separate. Drain acid phase away and discard.

24. Add 15 ml of 14N $HNO_3$ to the separatory funnel and equilibrate again for approximately 2-3 minutes; allow phases to separate. Drain acid phase away and discard.

25. Repeat step 24.

26. Remove yttrium from the organic phase by adding 15 ml of distilled water and equilibrating for approximately 2-3 minutes. Allow phases to separate and drain aqueous phase into 40-ml centrifuge tube.

27. Repeat step 26 using 15 ml of 0.1N $HNO_3$ instead of distilled water and drain acid phase into same 40-ml centrifuge tube as in step 26.

28. To precipitate yttrium oxalate, add 5 ml of 2N oxalic acid and adjust to pH 1.5 with approximately 2 ml of 15N $NH_4OH$ (using pH meter). Stir and cool in an ice bath approximately 20 minutes. Centrifuge and discard supernate.

29. Weigh planchet and 2.5-cm diameter membrane filter. Place membrane filter into filtering apparatus.

30. Take up precipitate in 10 ml of water and transfer to filtering apparatus. Wash with one 10-ml portion of distilled water, using the water to wash out centrifuge tube. Dry precipitate with three 20-ml portions of 95% ethyl alcohol and three 10-ml portions of diethyl ether. Continue suction for 2 minutes.

31. Transfer membrane filter with precipitate to stainless steel planchet and weigh after a delay of 10-15 minutes. Calculate the recovery of yttrium carrier.

32. Count the yttrium oxalate precipitate in a low-background beta counter (note b). Count again after 3 days to confirm absence of contamination of yttrium-90 by other radionuclides.

33. Dissolve strontium nitrate precipitate (obtained in step 21) in 10 ml of deionized distilled water and add 3 ml of 15N $NH_4OH$. Heat to near boiling in hot water bath and add 10 ml of 1N ammonium oxalate to precipitate strontium oxalate. Cool in ice bath, centrifuge, and decant.

34. Weigh a stainless steel planchet and 2.5-cm diameter membrane filter. Place filter into filtering apparatus.

35. Transfer the strontium oxalate precipitate to the filtering apparatus using three 10-ml portions of cold distilled water. Wash precipitate with three 10-ml portions of 95% ethyl alcohol and dry with three 10-ml portions of diethyl ether. Continue suction for 2 minutes.

36. Transfer the filter with precipitate to stainless steel planchet and weigh after a delay of 10-15 minutes. Calculate the recovery of strontium carrier (note c).

37. Count the precipitate for "total radiostrontium" activity.

Notes

a. Alternatively, the precipitate and filter paper may be dried at 100° C for 1 hour. After drying, cool in a desiccator 10-15 minutes. The recovery of barium carrier is then calculated from the weight of $BaCl_2$.

b. Care must be exercised that filter paper or membrane filter remains centered in planchet to avoid any geometric effects during counting.

c. Alternatively, the flame spectrophotometer may be used to determine recovery of strontium carrier by dissolving the precipitate after it has been counted. See section I, part E.

Calculation

Strontium-90: $$\text{pc/g ash} = \frac{(\text{net cpm})}{(A)(B)(C)(D)(E)(F)}$$

where (A) = recovery of strontium carrier,

(B) = recovery of yttrium carrier,

(C) = counting efficiency in $\frac{\text{cpm}}{\text{pc}}$ for yttrium-90 as yttrium oxalate mounted on a 2.5-cm membrane filter

(since 1 pc = 2.22 dpm, $\frac{\text{cpm}}{\text{pc}} = 2.22 \frac{\text{cpm}}{\text{dpm}}$),

(D) = weight of ash sample in grams,

(E) = correction factor for yttrium-90 decay ($e^{-\lambda t}$), where t is the time from the decantation of the yttrium-90 supernate to the time of counting,*

(F) = correction factor for the degree of equilibrium attained during the yttrium-90 ingrowth period ($1-e^{-\lambda t}$), where t is the time from start of the ingrowth period until the time of decantation of yttrium-90 supernate.**

$$\text{Strontium-89: pc/g ash} = \frac{1}{(A)(B)} \left[ \frac{(C)}{(D)(E)} - (F)(G)(H) - (G)(I)(J) \right]$$

where (A) = counting efficiency in $\frac{\text{cpm}}{\text{pc}}$ for strontium-89 as strontium oxalate mounted on a 2.5-cm diameter membrane filter

(since 1 pc = 2.22 dpm, $\frac{\text{cpm}}{\text{pc}} = 2.22 \frac{\text{cpm}}{\text{dpm}}$),

(B) = correction factor for strontium-89 decay ($e^{-\lambda t}$), where t is the time from sample collection to the time of counting,***

(C) = net cpm of "total radiostrontium",

(D) = recovery of strontium carrier,

(E) = weight of ash in grams,

(F) = self-absorption factor for strontium-90 as strontium oxalate mounted on 2.5-cm diameter membrane filter,

(G) = strontium-90 concentration in pc/g ash,****

(H) = counting efficiency in $\frac{\text{cpm}}{\text{pc}}$ for strontium-90 as strontium oxalate mounted on 2.5-cm diameter membrane filter,

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*Obtained from table 1, "Y-90 Decay and Ingrowth Factors", appendix B.
**Obtained from table 2, "Y-90 Ingrowth Factors", appendix B.
***Obtained from table 3, "Sr-89 Decay Factors", appendix B.
****Obtained from the calculation in section II, part B.

(I) = counting efficiency in $\frac{cpm}{pc}$ for yttrium-90 as yttrium oxalate mounted on 2.5-cm diameter membrane filter,

(J) = correction factor for yttrium-90 ingrowth $(1-e^{-\lambda t})$, where t is the time from the last decantation of $HNO_3$ from the strontium nitrate precipitate to the time of counting.*

Barium-140: $$pc/g\ ash = \frac{(net\ cpm)}{[(A)(B)(C)][(D) + (1.15)(E)(F)]}$$

where (A) = recovery of barium carrier,

(B) = weight of ash sample in grams,

(C) = correction factor for decay of barium-140 $(e^{-\lambda t})$, where t is the time from collection of the sample to the time of counting,**

(D) = counting efficiency in $\frac{cpm}{pc}$ for barium-140 on 2.4-cm filter paper

(since 1 pc = 2.22 dpm, $\frac{cpm}{pc} = 2.22\ \frac{cpm}{dpm}$),

(E) = counting efficiency in $\frac{cpm}{pc}$ for lanthanum-140 on 2.4-cm filter paper,

(F) correction factor for the ratio of lanthanum-140 activity to barium-140 activity $(1-e^{-0.015t})$, where t is the time in hours from the barium separation to the time of counting.***
The factor 1.15 in the denominator of the above equation is the ratio of lanthanum-140 activity to barium-140 activity in equilibrium.

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*Obtained from table 1, "Y-90 Decay and Ingrowth Factors", appendix B.
**Obtained from table 4, "Ba-140 Decay Factors", appendix B.
***Obtained from table 5, "La-140 Ingrowth Factors", appendix B.

## C. Calcium

### Principle of Method

An aliquot of an ashed sample is dissolved in hydrochloric acid. Calcium oxalate is precipitated from aliquots of this solution with oxalic acid, dissolved in dilute hydrochloric acid, and titrated against standardized potassium permanganate.

### Reagents*

Ammonium hydroxide, 6N
Ammonium oxalate, 0.03N
Hydrochloric acid, 6N
Oxalic acid, 1N
Potassium permanganate, 0.05N (note a)
Sodium oxalate standard

### Apparatus

Analytical balance
Burettes, 50 ml
Magnetic stirrer hot plate (stirring bars)
Whatman #542 filter paper, 11-cm diameter

### Procedure

1. Weigh accurately into a 40-ml centrifuge tube an appropriate amount (0.25 to 3 g, depending on calcium content) of sample ash (steps 1-5, sec. II, pt. A). RECORD WEIGHT OF ASH. Add 5 to 10 ml of 6N HCl and 10 ml of distilled water. Heat to dissolve as much solids as possible. Add 6N HCl to see if additional material will dissolve.

2. Transfer this solution quantitatively to a 50-ml volumetric flask. Dilute with distilled water to the mark. Shake well and filter the solution through a dry Whatman #542 filter paper fitted in a dry analytical funnel into a 125-ml Erlenmeyer flask.

3. Pipette two 20-ml aliquots of this solution into two 40-ml centrifuge tubes.

4. Heat in a water bath and add 10 ml of 1N oxalic acid to each and stir. Adjust each to pH 3 while hot with 6N $NH_4OH$ (use pH meter) to precipitate calcium oxalate. Cool slowly to room temperature, centrifuge, and discard the supernates.

*See Appendix A.

5. Wash each of the precipitates and the walls of each of the centrifuge tubes thoroughly, using not more than 10 ml of 0.03N ammonium oxalate. Centrifuge and discard the supernates.

6. Wash each of the precipitates with hot distilled water. Cool slowly to room temperature, centrifuge, and discard supernates (note b).

7. Dissolve each of the precipitates in approximately 2.5 ml of 6N HCl with gentle heating.

8. Dilute each to a volume of approximately 10 ml with distilled water. Transfer each quantitatively to 125-ml Erlenmeyer flasks, rinsing each of the centrifuge tubes with distilled water.

9. Add an additional 1 ml of 6N HCl and adjust the volume of each solution to approximately 25 ml with distilled water. Heat to just below boiling.

10. Titrate each while hot with standardized 0.05N $KMnO_4$ to the first faint pink end-point which persists for at least 30 seconds.

Notes

a. Standardize against NBS sodium oxalate standard. Weigh out in triplicate 80 mg of dry sodium oxalate standard. Dissolve each in 25 ml of $H_2O$ containing 2 ml of 6N HCl. Titrate each with approximately 0.05N $KMnO_4$ to a faint pink end-point which persists for at least 30 seconds. Standardize $KMnO_4$ at least once a week.

$$(\underline{N}\ KMnO_4) = \frac{g\ \text{sodium oxalate standard}}{(\text{ml of } KMnO_4)\ (0.067)}$$

b. It is important that all excess oxalic acid be removed from the precipitates. A stirring rod may be used to agitate the precipitates.

Calculation

$$g\ Ca/g\ ash = \frac{(\text{ml of } KMnO_4)\ \overline{(\underline{N}\ KMnO_4)}\ (\text{meq wt Ca})\ (\text{aliquot factor})}{(\text{wt of ash, g})}$$

where

(ml of $KMnO_4$) = volume of $KMnO_4$ solution used for titration,

$\overline{(N\ KMnO_4)}$ = average normality of $KMnO_4$ solution (average of three values from standardization),

(meq wt Ca) = milli-equivalent weight of calcium.

For the particular system described in the procedure, the equation reduces to

$$g\ Ca/g\ ash = \frac{(ml\ of\ KMnO_4)\ \ \overline{(N\ KMnO_4)}\ \ (0.05)}{(wt\ of\ ash,\ g)}$$

## D. Phosphate

### Principle of Method

The yellow molybdovanadophosphate complex, formed on addition of a mixed solution of sodium molybdate and sodium metavanadate to a sample solution containing phosphate, obeys Beer's Law in the range 0 to 50 $\mu$g/ml of phosphate. The intensity of the color formed is measured on a spectrophotometer at a wavelength of 434 m$\mu$. The transmittance of the sample is compared to a previously established calibration curve in order to determine the concentration of phosphate present.

### Reagents*

Hydrochloric acid, 6N (note a)
Mixed-color reagent (note a)
Phosphate standard solution (note a)

### Apparatus

Beckman Model DU spectrophotometer with tungsten filament lamp, blue filter, photomultiplier, and Sargent SR recorder
Silica cuvettes, matched
Whatman #542 filter paper, 11-cm diameter

*See appendix A.

Preparation of Calibration Curve

1. Pipette 1.0, 2.0, 3.0, 4.0, and 5.0 ml, respectively, of the phosphate standard solution to each of five 100-ml volumetric flasks.

2. Pipette 1.0 ml of 6N HCl into each volumetric flask.

3. Pipette 8.0 ml of mixed-color reagent into each flask, dilute to the mark with deionized distilled water, and mix thoroughly.

4. Reagent blank is prepared by pipetting 1.0 ml of 6N HCl and 8.0 ml of mixed-color reagent into a 100-ml volumetric flask, then diluting to the mark with deionized distilled water and mixing thoroughly.

5. Allow approximately 45 minutes for development of colored complex.

6. Transfer portions of the reagent blank and the dilutions of phosphate standard solution to cuvettes.

7. Determine the transmittance of the solutions on the spectrophotometer at a wavelength of 435 mμ (notes b and c).

8. Subtract the baseline response reading from standard response readings (note d). Plot on semilog paper with phosphate ion concentration as abscissa and relative response reading as ordinate.

Procedure

1. Transfer 0.2 to 0.3 g ash to a 250-ml Erlenmeyer flask; dissolve with 25 ml deionized distilled water and 25 ml of 6N HCl. Warm on hot plate for 20 minutes.

2. Filter sample through Whatman #542 filter paper into a 200-ml volumetric flask and wash residue thoroughly with deionized distilled water. After washing, dilute to 200 ml with deionized distilled water.

3. Pipette a 5.0-ml aliquot to a 100-ml volumetric flask. Pipette 8.0 ml of mixed color-reagent into flask, dilute to the mark with deionized distilled water, and mix thoroughly.

4. Allow approximately 45 minutes for color development.

5. Transfer sample to cuvettes.

6. Determine the transmittance on the Model DU spectrophotometer at a wavelength of 435 mμ.

7. Subtract baseline reading from sample reading (note d) and use calibration curve to determine concentration of phosphate ion.

Notes

a. All reagents and solutions for this procedure must be made with deionized distilled water.

b. Approximate settings of spectrophotometer used are:

| | |
|---|---|
| Wavelength | 435 mμ |
| Slit width | 0.04-0.05 mm |
| Sensitivity (power supply) | 7-9 |
| Blue filter | Middle position |
| Selector switch | 1 |
| Load resistor | 2 |
| Variable range (recorder) | 40-100 mv |

c. Adjust instrument settings so that reagent blank records at 90% of full scale and closed shutter records at 10% of full scale.

d. Shutter is closed between reading of each solution so that baseline fluctuations may be recorded.

## E. References

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## SECTION III. WATER

### A. Gross Alpha and Beta Activity

#### Principle of Method

The sample is acidified with hydrochloric acid and filtered. The suspended solids and the filter paper are ignited, the residue transferred to a planchet, and counted to determine gross alpha and beta activity. For treated water and rain samples, the filtrate is evaporated onto a planchet and the residue counted for gross alpha and beta activity of the dissolved solids. For untreated water, zinc, nickel, cobalt, iron, and strontium carriers are added to the filtrate. The basic sulfides are precipitated with hydrogen sulfide, filtered, ignited, and the residue transferred to a planchet and counted for alpha and beta activity. The filtrate from the sulfide precipitation is acidified and boiled to remove excess hydrogen sulfide. Strontium is precipitated from a basic solution as the carbonate and converted to the nitrate with concentrated nitric acid. The nitrate precipitate is dissolved, strontium carbonate is precipitated, collected by centrifugation, transferred to a planchet, dried, and counted for alpha and beta activity.

#### Reagents*

Ammonium hydroxide, 6N
Hydrochloric acid, 6N
Hydrogen sulfide, gas
Mixed carrier, 22 mg $Sr^{+2}$/ml and 5 mg/ml each of $Zn^{+2}$, $Fe^{+3}$, $Ni^{+2}$, and $Co^{+2}$
Nitric acid, 16N
Sodium carbonate, 3N
Sodium hydroxide, 6N

#### Apparatus

Analytical balance
Analytical oven

*See appendix A.

Desiccator
Infrared lamps
Muffle furnace
Stainless steel planchets, 2-in. diameter
Whatman #12 folded filter paper, 32-cm diameter
Whatman #541 filter paper, 12.5-cm diameter

## Procedure

1. Acidify the entire water sample (usually 1 gallon) with 2 ml of 6N HCl. Filter a 1-liter aliquot of the sample through a Whatman #12 folded filter paper. Wash filter paper with approximately 100 ml of distilled water. Combine the filtrate and washing. If the sample is salt water, the filter paper should be washed with an additional 500 ml of warm distilled water. This second wash is combined with the filtrate.

## Suspended Solids

2. Dry the filter and suspended solids and ignite for 1-2 hours in a muffle furnace at 550° C. Cool.

3. Transfer residue with distilled water to a weighed, stainless steel planchet. Dry in oven at 110° C for 2 hours and cool in desiccator for 1 hour (until constant weight). Weigh sample and planchet. Store sample in desiccator (silica gel desiccant) until sample is to be counted. Count for alpha and beta activity. This planchet is designated as "suspended solids". If the sample is treated water or rain water, proceed to step 4; otherwise, proceed to step 7.

## Dissolved Solids (treated water or rain)

4. The filtrate from step 1 is evaporated to approximately 75 ml and transferred with distilled water to a 100-ml beaker; continue the evaporation. At this time, some of the filtrate can be added to a weighed stainless steel planchet and evaporated under infrared lamp. Continue evaporation until entire sample has been transferred to the planchet and dried.

5. Dry in oven at 110° C for 2 hours and cool in desiccator for 1 hour (until constant weight). Weigh sample and planchet. Store samples in desiccator (silica gel desiccant) until sample is to be counted.

6. Count to determine alpha and beta activities.

Dissolved Solids (untreated water with high dissolved-solids content)

Sulfide Precipitation

7. To the filtrate from step 1, add 2 ml of mixed carrier. Acidify the solution with 2 ml of 6N HCl.

8. Heat solution to boiling. Adjust to pH 8 (pH paper) with 6N $NH_4OH$.

9. Precipitate with $H_2S$ gas, stirring vigorously. Continue to stir regularly for 10-15 minutes, and digest for 1 hour, maintaining pH 8 with 6N $NH_4OH$.

10. Cool, filter the solution through a Whatman #541 filter paper, and wash precipitate with approximately 100 ml of hot distilled water. Save the filtrate for use at step 14.

11. Dry and ignite the precipitate and paper at 550° C for 1 hour.

12. Wash the residue from the ignition crucible into a weighed planchet.

13. Dry the residue under an infrared lamp and weigh. Designate as "basic sulfides". Count to determine alpha and beta activity.

Carbonate Precipitation

14. The filtrate from step 10 is acidified with 2 to 3 ml of 6N HCl. Boil solution for 2-3 hours to remove excess $H_2S$, adding distilled water to keep the volume at approximately 1 liter. Filter the solution and discard the filter.

15. Heat the filtrate to boiling, adjust to pH 10 (pH paper) with 6N NaOH, and add, with stirring, 50 ml of 3N $Na_2CO_3$. Continue heating until precipitate settles to the bottom of the beaker. Check for completeness of precipitation with a few drops of 3N $Na_2CO_3$. Remove from hot plate and let solution cool to room temperature.

16. Decant supernate. Transfer the precipitate to a 250-ml centrifuge bottle, wash with 50 ml of distilled water, and centrifuge. Decant and discard the supernate.

17. Dissolve the precipitate in 100 ml of 6N HCl, transfer the solution to a 1-liter beaker, and heat to boiling. Add, with stirring,

110 to 115 ml of 6N $NH_4OH$ to make the solution basic. Add 30 ml of 3N $Na_2CO_3$ to precipitate the carbonates. Check for completeness of precipitation with a few drops of 3N $Na_2CO_3$.

18. Allow the solution to cool and decant the bulk of the supernate. Transfer the carbonate precipitate to a 250-ml centrifuge bottle, centrifuge, and decant supernate. Wash with 50 ml of distilled water, centrifuge, decant, and discard supernate. Dry precipitate at 100° C for 5 hours or longer.

19. Remove from oven, cool, and dissolve precipitate in approximately 10 ml of 6N $HNO_3$ (warming in a water bath with occasional stirring may aid dissolution). Transfer solution to a 40-ml graduated centrifuge tube. Add a sufficient volume of 90% $HNO_3$ to obtain a 70% nitric acid concentration (table 1-1). Cool in an ice bath approximately 10 minutes, centrifuge, and discard the supernate.

20. To the precipitate, add 30 ml of 16N $HNO_3$, heat in hot water bath for 10-15 minutes, cool in ice bath, and centrifuge to collect nitrate precipitate. Decant and discard supernate.

21. Dissolve the nitrate precipitate in 10 ml of distilled water. Adjust solution to pH 9 (pH paper) with 6N $NH_4OH$ and add 10 ml of 3N $Na_2CO_3$ to precipitate carbonates.

22. Centrifuge, decant, and discard supernate. Transfer carbonate precipitate to a weighed stainless steel planchet using distilled water. Dry precipitate under infrared lamp and weigh. Designate as "carbonates". Count to determine alpha and beta activity.

Calculation

Gross Beta:
$$pc/l = \frac{(\text{net beta cpm})}{(A)(B)(C)}$$

where (A) = counting efficiency in $\frac{cpm}{pc}$ for gross beta activity (calibrated with equilibrium mixture of strontium-90 + yttrium-90)

(since 1 pc = 2.22 cpm, $\frac{cpm}{pc} = 2.22 \frac{cpm}{dpm}$),

(B) = volume of sample in liters,

(C) = correction factor for self-absorption in the sample.

Gross Alpha: $$pc/l = \frac{(\text{net alpha cpm})}{(A)(B)(C)}$$

where (A) = alpha counting efficiency in $\frac{cpm}{pc}$ for a "weightless" sample,

(B) = volume of sample in liters,

(C) = correction factor for self-absorption in the sample.

## B. Strontium-90, Strontium-89, and Barium-140

### Principle of Method

Strontium and barium, as carriers added to an aliquot of the filtered water sample, are precipitated as carbonates, then converted to the nitrates. The nitrate precipitate is dissolved in distilled water and the solution scavenged by precipitation of mixed hydroxides with dilute ammonium hydroxide. Barium is precipitated as the chromate, purified as the chloride, then counted to determine barium-140. Strontium is precipitated from the supernate as the carbonate, converted to the nitrate, and stored for yttrium-90 ingrowth. The strontium nitrate is dissolved, yttrium carrier added, and strontium reprecipitated with concentrated nitric acid. Yttrium is extracted from the supernate with tributyl phosphate, reextracted with dilute nitric acid, and precipitated as the oxalate. The yttrium oxalate is weighed to determine recovery of yttrium carrier, then counted. The strontium nitrate is dissolved in deionized distilled water and reprecipitated as the nitrate. The nitrate is dissolved and precipitated as strontium oxalate. The oxalate precipitate is filtered, weighed to determine strontium carrier recovery, and counted for "total radiostrontium". The strontium-89 activity is calculated from the difference between the count for "total radiostrontium" and the count due to the previously determined strontium-90 activity.

### Reagents*

Ammonium acetate buffer, pH 5
Ammonium hydroxide, 15N, 6N
Ammonium oxalate, 1N
Barium carrier, 20 mg $Ba^{+2}$/ml
Diethyl ether

*See appendix A.

Ethyl alcohol, absolute, 95%
Hydrochloric acid, 12N
Hydrochloric acid-diethyl ether reagent
Mixed rare earth carrier
Nitric acid, 90%, 16N, 14N, 8N, 6N, 0.1N
Oxalic acid, 2N
Sodium carbonate, 3N, 0.1N
Sodium chromate, 3N
Sodium hydroxide, 6N
Strontium carrier, 20 mg $Sr^{+2}$/ml
Tributyl phosphate (TBP), preequilibrated
Yttrium carrier, 10 mg $Y^{+3}$/ml

## Apparatus

Analytical balance
Analytical oven
Desiccator
Infrared lamp
Millipore filtering apparatus
Millipore #OH membrane filter (1.5-$\mu$ pore size), 2.5-cm diameter
pH meter
Stainless steel planchets, 2-in. diameter
Whatman #12 folded filter paper, 32-cm diameter
Whatman #542 filter paper, 2.4-cm diameter

## Procedure

1. Acidify the sample (usually 1 gallon) with 2 ml of 6N HCl. Mix thoroughly and filter through Whatman #12 folded filter paper. Place a 1-liter aliquot of the filtered sample in a 2-liter beaker. Add 1.0 ml of strontium carrier and 1.0 ml of barium carrier, stir well, and heat to near boiling on hot plate.

2. Adjust to pH 10 (pH paper) with 10 ml of 6N NaOH. Add 30 ml of 3N $Na_2CO_3$ with stirring. Continue heating until precipitate settles to the bottom of the beaker. Remove from hot plate and let solution cool to room temperature.

3. Decant supernate. Transfer precipitate to a 40-ml graduated centrifuge tube with distilled water. Centrifuge and discard the supernate. Add 20 ml of 0.1N $Na_2CO_3$ to the residue, stir, centrifuge, and discard supernate.

4. Dissolve the residue in 5 ml of 6N $HNO_3$. Add 25 ml of 90% $HNO_3$ with stirring. Cool in ice bath for 10 minutes. Centrifuge and discard supernate.

5. Add 30 ml of 16N $HNO_3$, stir, cool in ice bath for 10 minutes, centrifuge, and discard the supernate.

6. Dissolve the nitrate precipitate in 20 ml of distilled water. Add 1 ml of mixed rare earth carrier and adjust solution to pH 7 (pH paper) with 6N $NH_4OH$. Heat, centrifuge, save supernate, and discard precipitate.

7. To the supernate add 5 ml of ammonium acetate buffer and heat in water bath. Check for pH 5 with pH paper. Add additional buffer solution if necessary. To precipitate $BaCrO_4$, heat solution in a water bath and add 1 ml of 3N $Na_2CrO_4$ dropwise with stirring. Centrifuge and decant the supernate into a clean 40-ml centrifuge tube. If barium-140 analysis is desired, continue working with precipitate and follow steps 8-13; otherwise, continue working with supernate at step 14.

8. Add approximately 5 ml of 12N HCl to the $BaCrO_4$ precipitate. Heat in a water bath until the yellow $BaCrO_4$ precipitate is dissolved and a white granular $BaCl_2.2H_2O$ precipitate remains. At this point the solution will have a green coloration from $Cr^{+3}$.

9. Cool in ice bath and add 20 ml of cold HCl-diethyl ether reagent. Stir, centrifuge, and decant the supernate. A pure white precipitate ($BaCl_2.2H_2O$) should remain. If a coloration occurs, repeat the HCl-diethyl ether application. RECORD THE TIME OF DECANTATION AS THE START OF LANTHANUM-140 INGROWTH.

10. Weigh a stainless steel planchet and 2.4-cm diameter Whatman #542 filter paper. Place filter paper into Millipore filtering apparatus.

11. Transfer the precipitate onto the filter paper with several 10-ml portions of cold absolute ethyl alcohol. Use at least 30 to 40 ml of absolute ethyl alcohol.

12. Transfer filter paper with precipitate to stainless steel planchet. Allow precipitate to remain in contact with the atmosphere until constant weight is achieved (approximately 2 hours). Weigh and calculate the recovery of barium carrier as $BaCl_2.2H_2O$.

13. Count the precipitate on the planchet in a low-background beta counter (note b).

14. To the supernate from step 7, add sufficient 3N $Na_2CO_3$ (at least 5 ml) to precipitate strontium carbonate. Allow solution to stand for a few minutes. Stir, centrifuge, and check supernate

for completeness of precipitation with a few drops of 3N $Na_2CO_3$. Decant the supernate. Wash the strontium carbonate precipitate with 20 ml of 0.1N $Na_2CO_3$. Centrifuge and decant the supernate.

15. Dissolve precipitate in 5 ml of 6N $HNO_3$. Add 25 ml of 90% $HNO_3$, stir, and cool in ice bath for 10 minutes. Centrifuge and decant supernate. RECORD TIME OF DECANTATION AS START OF YTTRIUM-90 INGROWTH. Drain excess acid on paper towels.

16. Store strontium nitrate precipitate for 2 weeks to allow ingrowth of yttrium-90.

17. After the ingrowth period, add 1.0 ml of yttrium carrier and 1.0 ml of 8N $HNO_3$ to the centrifuge tube and warm gently to dissolve precipitate. Add 10 ml of 90% $HNO_3$, stir, and cool in ice bath for 10 minutes. Centrifuge and quantitatively decant the supernate into a 60-ml separatory funnel. RECORD THE TIME OF SEPARATION AS START OF YTTRIUM-90 DECAY. Save the precipitate (for step 28). Drain excess $HNO_3$ from centrifuge tube by inverting tube on a paper towel.

18. To the solution in the separatory funnel, add 4 ml of distilled water to make the solution 14N in nitric acid. Add 10 ml of preequilibrated TBP and equilibrate by shaking for 2-3 minutes. Allow phases to separate, then drain acid phase away and discard.

19. Add 15 ml of 14N $HNO_3$ to the separatory funnel and equilibrate again for 2-3 minutes. Allow phases to separate, drain acid phase away, and discard.

20. Repeat step 19.

21. Remove yttrium from the organic phase by adding 15 ml of distilled water and equilibrating for 2-3 minutes. Allow phases to separate and drain aqueous phase into 40-ml centrifuge tube.

22. Repeat step 21, using 15 ml of 0.1N $HNO_3$ instead of distilled water, and drain acid phase into the same 40-ml centrifuge tube used in step 21.

23. To precipitate yttrium oxalate, add 5 ml of 2N oxalic acid and adjust to pH 1.5 with approximately 2 ml of 15N $NH_4OH$ (using pH meter). Stir and cool in an ice bath for approximately 20 minutes. Centrifuge and discard supernate.

24. Weigh planchet and 2.5-cm diameter membrane filter. Place membrane filter into filtering apparatus.

25. Take up precipitate in 10 ml of cold water and transfer to filtering apparatus. Wash with one 10-ml portion of cold distilled water, using the water to wash out centrifuge tube. Dry precipitate with three 20-ml portions of 95% ethyl alcohol and three 10-ml portions of diethyl ether. Continue suction for 2 minutes.

26. Transfer membrane filter with precipitate to stainless steel planchet and weigh after a delay of 10-15 minutes. Calculate the recovery of yttrium carrier.

27. Count the yttrium oxalate precipitate in a low-background beta counter (note b). Count again after 3 days to confirm absence of contamination of yttrium-90 by other radionuclides.

28. Dissolve the strontium nitrate precipitate (obtained in step 17) in 10 ml of deionized distilled water. Add 30 ml of 90% $HNO_3$, stir, cool in ice bath, centrifuge, and decant. RECORD THE TIME OF DECANTATION AS START OF YTTRIUM-90 INGROWTH.

29. Dissolve strontium nitrate precipitate in 10 ml of deionized distilled water and add 3 ml of 15N $NH_4OH$. Heat to near boiling in hot water bath and add 5 ml of 1N ammonium oxalate to precipitate strontium oxalate. Cool in ice bath, centrifuge, and decant.

30. Weigh a stainless steel planchet and 2.5-cm diameter membrane filter. Place filter into filtering apparatus.

31. Transfer the strontium oxalate precipitate to the filtering apparatus using three 10-ml portions of cold distilled water. Wash precipitate with three 10-ml portions of 95% ethyl alcohol and dry with three 10-ml portions of diethyl ether. Continue suction for 2 minutes.

32. Transfer the filter with precipitate to stainless steel planchet and weigh after a delay of 10-15 minutes. Calculate the recovery of strontium carrier (note c).

33. Count the precipitate for "total radiostrontium" activity.

Notes

a. Alternatively, the precipitate is dried at 100° C for 1 hour, following the final absolute ethyl alcohol treatment. The recovery of barium carrier is then calculated from the weight of $BaCl_2$.

b. Care must be exercised that filter paper or membrane filter remains centered in planchet to avoid any geometric effects during counting.

c. Alternatively, the flame spectrophotometer may be used to determine recovery of strontium carrier by dissolving the precipitate after it has been counted. See section I, part E.

Calculation

Strontium-90: $$pc/l = \frac{(\text{net cpm})}{(A)(B)(C)(D)(E)(F)}$$

where (A) = recovery of strontium carrier,

(B) = recovery of yttrium carrier,

(C) = counting efficiency in $\frac{cpm}{pc}$ for yttrium-90 as yttrium oxalate mounted on a 2.5-cm membrane filter

(since 1 pc = 2.22 dpm, $\frac{cpm}{pc} = 2.22 \frac{cpm}{dpm}$),

(D) = sample volume in liters,

(E) = correction factor for yttrium-90 decay ($e^{-\lambda t}$), where t is the time from the decantation of the yttrium-90 supernate to the time of counting,*

(F) = correction factor for the degree of equilibrium attained during the yttrium-90 ingrowth period ($1-e^{-\lambda t}$), where t is the time from the start of the ingrowth period until the time of decantation of the yttrium-90 supernate.**

Strontium-89: $$pc/l = \frac{1}{(A)(B)} \left[ \frac{(C)}{(D)(E)} - (F)(G)(H) - (G)(I)(J) \right]$$

where (A) = counting efficiency in $\frac{cpm}{pc}$ for strontium-89 as strontium oxalate mounted on a 2.5-cm diameter membrane filter

(since 1 pc = 2.22 dpm, $\frac{cpm}{pc} = 2.22 \frac{cpm}{dpm}$),

*Obtained from table 1, "Y-90 Decay and Ingrowth Factors", appendix B.
**Obtained from table 2, "Y-90 Ingrowth Factors", appendix B.

(B) = correction factor for strontium-89 decay ($e^{-\lambda t}$), where t is the time from sample collection to the time of counting,*

(C) = net cpm of "total radiostrontium".

(D) = recovery of strontium carrier,

(E) = volume of sample in liters,

(F) = self-absorption factor for strontium-90 as strontium oxalate mounted on 2.5-cm diameter membrane filter,

(G) = strontium-90 concentration in pc/l, **

(H) = counting efficiency in $\frac{\text{cpm}}{\text{pc}}$ for strontium-90 as strontium oxalate mounted on 2.5-cm diameter membrane filter,

(I) = counting efficiency in $\frac{\text{cpm}}{\text{pc}}$ for yttrium-90 as yttrium oxalate mounted on 2.5-cm diameter membrane filter,

(J) = correction factor for yttrium-90 ingrowth ($1-e^{-\lambda t}$), where t is the time from the second decantation of $HNO_3$ from the strontium nitrate precipitate to the time of counting.***

Barium-140:
$$\text{pc/l} = \frac{(\text{net cpm})}{[(A)(B)(C)]\ [(D) + (1.15)(E)(F)]}$$

where (A) = recovery of barium carrier,

(B) = volume of sample in liters,

(C) = correction factor for decay of barium-140 ($e^{-\lambda t}$), where t is the time from collection of the sample to the time of counting,****

*Obtained from table 3, "Sr-89 Decay Factors", appendix B.
**Obtained from the calculation in section III, part B.
***Obtained from table 1, "Y-90 Decay and Ingrowth Factor", appendix B.
****Obtained from table 4, "Ba-140 Decay Factors", appendix B.

(D) = counting efficiency in $\frac{cpm}{pc}$ for barium-140 on 2.4-cm filter paper

(since 1 pc = 2.22 dpm, $\frac{cpm}{pc} = 2.22 \frac{cpm}{dpm}$),

(E) = counting efficiency in $\frac{cpm}{pc}$ for lanthanum-140 on 2.4-cm filter paper,

(F) = correction factor for the ratio of lanthanum-140 activity to barium-140 activity ($1-e^{-0.015t}$), where t is the time in hours from the barium separation to the time of counting,*

The factor 1.15 in the denominator of the above equation is the ratio of lanthanum-140 activity to barium-140 activity in equilibrium.

## C. Tritium

### Principle of Method

The water sample is distilled and a portion of the distillate is pipetted into a counting vial containing the dioxane scintillation mix. After mixing, the contents of the vial are counted in a liquid scintillation counter.

### Reagents

Scintillation mix, dioxane

### Apparatus

Burette, 100 ml
Condenser, miniature
Distillation flask, 50 ml
Pipettes, 3 ml
Polyethylene liquid scintillation counting vials

*Obtained from table 5, "La-140 Ingrowth Factors", appendix B.

## Procedure

1. Distill a water sample (10 to 50 ml) to dryness (note a), collecting the distillate in a small container.

2. Dispense 13.0 ml of the scintillation mix from a burette into a polyethylene counting vial (note b).

3. Pipette 3.0 ml of the water distillate into the vial and mix contents with gentle swirling (note c).

4. Prepare a background sample from distilled water in the same manner as described in steps 2 and 3 (note d).

5. Prepare a counting standard, using a tritiated water standard in the same manner as described in steps 2 and 3 (note e).

6. Place samples in freezer section of counter for at least 30 minutes before commencing to count.

## Notes

a. Water sample should be distilled to dryness. Baking of the residue should be avoided, however, so that other fission-product activities will not be distilled as well.

b. Do not label vials; only the caps.

c. Avoid direct sunlight and use a minimum of artificial light during sample preparation.

d. It is preferable to use water known to have low tritium content, e.g., triple distilled deep sea water or deep well water.

e. Commercial standards are most conveniently diluted to approximately $3 \times 10^4$ dpm/ml.

## Calculation

Tritium:

$$pc/l = \frac{1000\,(A)}{(B)(C)}$$

where (A) = net cpm,

(B) = volume of water taken for sample (3.0 ml),

(C) = counting efficiency in cpm/pc for tritium

(since 1 pc = 2.22 dpm, $\frac{cpm}{pc} = 2.22\,\frac{cpm}{dpm}$).

## D. References

Association of Official Agricultural Chemists: "Determination of Phosphorus", Official Methods of Analysis, 9th ed. (1960) p. 9.

Goldin, A. S., and R. J. Velten: "Application of Tributyl Phosphate Extraction to the Determination of Strontium-90", Analytical Chemistry 33:149 (1961).

Sodd, V. J., A. S. Goldin, and R. J. Velten: "Determination of Radioactivity in Saline Waters", Analytical Chemistry 32:25 (1960).

American Public Health Association: "Strontium-90 in Water", Standard Methods for the Examination of Water and Wastewater, 11th ed., p. 451.

## SECTION IV. COUNTING PROCEDURES

### A. Low-Background Beta Counting

#### Instrumentation

The beta counters are of the low-background, multiple-detector type with two to four sample detectors per unit. Low background is achieved through the use of steel or lead shielding plus guard counters operating in anticoincidence with the sample detectors. The sample detectors are gas-flow, window-type Geiger counters, approximately 2 inches in diameter. The counting gas is a mixture consisting of 99.05% helium and 0.95% isobutane. The beta counters are equipped with preset timers. All samples are placed in 2-in. diameter stainless steel planchets with 1/4-in. walls for counting.

#### Operating Voltage

The voltage on the detector necessary for proper operation is determined by running the "plateau curve" of count rate versus high voltage. The correct operating voltage is selected as the point approximately one-third of the distance from the beginning of the plateau to the end of the plateau. The correct operating voltage should be determined at least once each month and, in addition, after the gas supply is changed or after any modification is made on a detector. Any appreciable change in the operating voltage, plateau length, or plateau slope is an indication of possible trouble in a detector or in the counting gas. When the counting gas is changed on a counter, be sure to purge the detector for several hours before using it.

#### Counting Efficiency

Counting efficiency is the relationship between sample disintegration rate and sample counts recorded by the counter. Counting efficiency is usually expressed as count/disintegration or as the percentage of disintegrations recorded as counts. Counting efficiency may also be expressed as the count rate per unit of radioactivity, i.e., $\frac{\text{cpm}}{\text{pc}}$.

Counting efficiency is determined by placing a source with a known disintegration rate or known radioactivity in the beta

counter and measuring its count rate. This source is referred to as a standard source and the process of determining the counting efficiency is called calibration. Calibration should be performed for each radionuclide that will be counted in the counter, and the standard sources should be of the same physical form (size, shape, planchet material, etc.) as the samples to be counted. Gross beta calibration is performed using a standard source of strontium-90 plus yttrium-90 in equilibrium. The result of a gross beta determination of a sample is then reported as activity equivalent to strontium-90 plus yttrium-90 in equilibrium.

Beta counters should be calibrated every 4-6 months and after any mechanical modifications are made on a detector. The daily operational check procedure as outlined below serves also as check on the relative counting efficiency.

Background Determination

The background counting rate of the low-background beta counters runs from 0.5 to 1.5 counts per minute and varies from detector to detector and from day to day. The background for each detector should be determined at the beginning of the workday and should be measured using a clean, empty planchet in each counter.

Absorption Factor

The beta counting efficiency is determined using an essentially weightless standard source. In many types of samples there is a sufficient amount of solids so that the sample partially shields itself and produces "self-absorption". In this case, the count rate obtained on the sample is not as great as it should be and must be corrected for self-absorption according to the weight, or more correctly the "density-thickness" ($mg/cm^2$), of the sample.

The absorption correction factors should be determined for the specific radionuclide being counted and for approximately the same chemical composition as in the actual samples. A curve of the absorption correction factor versus sample weight or density-thickness can be obtained by making up samples containing equal concentrations of radioactivity per unit weight of solids and varying amounts of solids (solids content should vary from approximately zero solids to a weight equal to the maximum weight expected in an actual sample). Be sure that the radioactivity is uniformly distributed in the solid material. The count rate per unit weight of solids for each of the samples is then divided by the count rate of the weightless sample, and this fraction is plotted as a function of the sample weight. It is not necessary to use a calibrated solution for this study since all values are compared

to the weightless sample value. For the gross beta absorption factor determination one should use strontium-90 plus yttrium-90 (in equilibrium) activity.

## Sample Handling

All samples should be handled with care to prevent accidental loss of the material in the planchet, contamination of the counting room, or contamination of the beta counters. All samples should be kept covered when not actually in a counter to prevent dust from getting into the samples. Samples which tend to be hygroscopic should be kept in a desiccator until counted. Planchets that have material piled above the top edge of the wall or have material on the outside of the planchet should not be placed in the beta counters. Return these samples to the laboratory for repacking. Some types of samples consist of a precipitate on a 2.5-cm diameter filter paper. The filter paper should always be in the center of the 2-in. planchet when the planchet is being placed into the counter.

## Counter Operation Check

At the beginning of each workday, a standard check source should be counted to determine that the counter is operating satisfactorily. The count obtained each day should be compared with the usual count of the check source for that particular detector. A count significantly higher or lower than the usual value is an indication of detector malfunction or insufficient counting-gas supply. A graph of the daily check source counts showing the acceptable limits of variation is desirable.

## Sample Types and Counting Times

Table 4-1 gives the types of samples counted for beta radioactivity, counting times, and general remarks pertaining to each type.

Table 4-1. Beta Counting Sample Types and Counting Times

| Sample type | Nuclide | Counting time | Remarks |
|---|---|---|---|
| a. Yttrium oxalate on 2.5-cm diameter filter paper (milk, water, food, biota, silt, and soil) | Y-90 | 50 min | Place filter in center of 2-in. planchet. Count as soon as possible to reduce correction for Y-90 decay. |
| b. Strontium oxalate (total strontium) on 2.5-cm diameter filter paper (milk, water, food, biota, silt, and soil) | Sr-89 & Sr-90 | 50 min | Place filter in center of 2-in. planchet. Count as soon as possible to reduce correction for Y-90 ingrowth and Sr-89 decay. |
| c. Barium chloride on 2.4-cm diameter filter paper (milk, water, food, biota, silt, and soil) | Ba-140 | 50 min | Place filter in center of 2-in. planchet. |
| d. Gross beta 2-in. planchet (water, food, biota, silt, and soil) | Gross beta | 20-50 min, depending on count rate of sample | Handle samples carefully to avoid spilling contents. |

## B. Alpha Counting

### Instrumentation

The alpha counters are the internal proportional gas-flow type and are operated on the alpha plateau in the proportional region. The counting gas is "P-10", a mixture of 90% argon and 10% methane. The detectors are hemispherical with approximately 2.5-in. diameters. After insertion of each sample, the detector must be purged for approximately 3 minutes before the count is started.

### Operating Voltage

The plateau curves for the alpha counters are run in the same manner as is described in part A above. In addition, to assure that the counter is on the alpha plateau rather than the alpha and beta plateau, a beta standard is counted in the chamber. The voltage on the detector is lowered until it counts only alpha disintegrations and the alpha plateau curve is run in this region. The correct operating voltage is taken as the midpoint of the alpha plateau.

Counting Efficiency

The standards used in determining alpha counting efficiencies should approximate the samples to be counted in physical size and backing material. Again, comments found in part A apply here. Natural uranium (in equilibrium) or a specific alpha emitter such as plutonium-239 may be used as the calibration source.

Background Determination

The background counting rate of the alpha counters varies from 0.12 to 0.18 count per minute. Each day before any samples are counted, a 100-minute background for each detector is determined with an empty planchet in place. If the background is above 0.18 count per minute, the chamber is decontaminated as outlined below.

Self-Absorption Factor

Because an alpha particle is readily stopped in matter, the number reaching the sensitive volume of the detector is greatly reduced by self-absorption and scattering in the sample material. To correct for this, it is necessary to determine the self-absorption factors for alpha counting. Alpha self-absorption factors are determined in the same manner as the beta self-absorption factors. Samples are prepared containing equal concentrations of alpha radioactivity per unit weight and varying amounts of solids, and the count per unit weight is plotted as a function of the solids weight (mg) or "density-thickness" ($mg/cm^2$). The samples used to determine self-absorption should resemble the actual samples as much as possible in physical size, planchet material, and chemical composition, and the alpha emitter should be the same as used to determine alpha counting efficiency.

As it is usually not practical to prepare a "weightless" alpha sample in the laboratory, the self-absorption curve may be extrapolated to find the count rate at zero density-thickness. If a standard solution is used to prepare the self-absorption samples, the "weightless" sample counting efficiency is determined from the extrapolated count rate at zero density-thickness. The self-absorption factors are computed by dividing each count rate per unit weight by the extrapolated "weightless" sample count rate. The self-absorption factors thus computed may be plotted as a function of either total sample weight (mg) or density-thickness ($mg/cm^2$) to obtain the self-absorption curve.

Sample Handling

The same comments apply here as given in part A.

Counter Operation Check

The same procedure is followed here as outlined in part A.

Decontamination

Because the alpha samples are placed inside the detector, an internal proportional detector may be easily contaminated. Approximately every 3 months, or at any time the background increases above the level normally encountered, the detector should be removed and thoroughly cleaned with ethyl alcohol. A brown film often appears on the walls of the detector. This film should be removed by lightly scrubbing with scouring powder. Be careful to remove only the brown film and not the conductive coating inside the chamber. The center wire should be cleaned by dipping in ethyl alcohol (with care not to bend it out of its normal shape). The accumulation of this deposit on the chamber walls leads to a gradual deterioration in performance (generally observed as an increasing operating voltage, change in plateau slope, and decreasing plateau length). A sudden deterioration in performance is usually indicative of mechanical damage to the center wire or an accumulation of dust or debris on the center wire.

Sample Types and Counting Times

Table 4-2 gives the types of samples counted for alpha activity, counting times, and general remarks pertinent to each sample type.

Table 4-2. Alpha Counting Sample Types and Counting Times

| Sample type | Nuclide | Counting time | Remarks |
|---|---|---|---|
| a. Gross alpha, 2-in. planchet (water, food, biota, silt, and soil) | Gross alpha | 25-100 min | Handle planchet carefully to avoid spilling contents. |
| b. Barium sulfate (total radium) on 2.5-cm diameter filter paper (water, food, biota, silt, and soil) | "Total radium" | 100 min | Place filter in center of 2-in. planchet. Count as soon as possible to avoid correction for radium daughter ingrowth. |

## C. Gamma Spectroscopy

Sample Preparation

All supplies are usually gamma scanned for specific gamma-emitting nuclides before any processing is done on the sample, i.e., they are gamma scanned in the "raw" form as received at the laboratory. The only preparation required is to make sure the sample is homogeneous. For solid media it is necessary to grind and blend the sample before counting. A 3.5-liter aliquot of the

homogeneous sample is placed in a clean, polyethylene, Marinelli-type beaker and analyzed by gamma spectroscopy.

Instrumentation

The gamma spectrometer is comprised of a detector, multichannel pulse height analyzer, data read-out device, and auxiliary instrumentation. The detectors are 4- by 4-in. NaI(Tl) crystals coupled to photomultiplier tubes and enclosed in steel cave-type shields. The Marinelli-type beaker containing the 3.5-liter sample is placed over the detector inside of the shield.

Calibration

Two types of calibration are required in gamma spectroscopy: (1) gamma energy vs. channel number; and (2) gamma-counting efficiency. Knowledge of the energy-channel number relationship permits qualitative identification of specific gamma-emitting nuclides. In order to make a quantitative determination of the amount of a particular nuclide, it is necessary to know the counting efficiency for that nuclide.

(1) Gamma Energy-Channel Number Relationship: The spectrometers are used with two energy-channel number relationships. For most of the samples from routine programs, 100 channels of an analyzer are used to cover the energy range from 0 to 2 Mev, or 20 Kev per channel. For some programs, however, 200 channels are used for an energy range of 0 to 2 Mev, or 10 Kev per channel. The spectrometers are aligned to the proper energy-channel number relationship by counting several nuclides with gamma energies in the energy range of interest (0 to 2 Mev) and plotting the location (channel number) of each peak as a function of the known gamma-ray energy. Varying the gain of the amplifier in the analyzer changes the slope of this line and, in effect, changes the energy-channel number relationship by a constant factor (multiplier). Increasing the gain by a factor of 2X changes the slope by a factor of 2X and changes the energy calibration by a factor of 2X. Thus, a peak which initially was located in channel number 10 would now appear in channel number 20. This relationship is true only if the plot of channel number versus gamma energy passes through the origin (zero). The threshold control changes the X-axis intercept and permits one to adjust the spectrometer so that the line passes through the origin. Often it will be necessary to adjust the threshold, adjust the gain, readjust the threshold, and readjust the gain, etc., to achieve perfect calibration. Table 4-3 shows the nuclides which are used for energy calibration and/or efficiency calibration.

(2) Gamma Counting Efficiency: The counting efficiency should be determined for each of the nuclides which one desires to quantitatively measure. Counting efficiency is usually expressed in units of counts per minute in a selected spectral

region per picocurie ($\frac{cpm}{pc}$) or counts per minute per picocurie per liter ($\frac{cpm}{pc/l}$) A known amount of activity of a particular nuclide is diluted to a 3.5-liter volume with distilled water, thoroughly mixed, placed in a Marinelli beaker, and counted on the spectrometer. This standard solution should be counted for a sufficient length of time to obtain a total count which will yield good counting statistics. A total count (in the spectral region of interest) of approximately 10,000 counts is usually adequate.

Table 4-3. Nuclides Useful for Calibrating Spectrometer

| Nuclide | Half-life* | Energy of principle gamma rays (Mev) | Channel No. | |
|---|---|---|---|---|
| | | | 10 Kev | 20 Kev |
| Ce-Pr-144 | 285 d | 0.134 | 13 | 7 |
| I-131 | 8.08 d | 0.364 | 36 | 18 |
| | | 0.637 | 64 | 32 |
| Ru-Rh-106 | 1.00 y | 0.513 | 51 | 25 |
| Cs-137 | 26.6 y | 0.662 | 66 | 33 |
| Zr-Nb-95 | 65 d | 0.757 | 76 | 38 |
| | | 0.724 | 72 | 36 |
| | | 0.765 | 76 | 38 |
| Zn-65 | 245 d | 1.119 | 112 | 56 |
| K-40 | $1.39 \times 10^9$ y | 1.46 | 146 | 73 |
| Ba-La-140 | 12.80 d | 0.487 | 49 | 25 |
| | | 0.537 | 54 | 27 |
| | | 1.596 | 160 | 80 |
| Ru-Rh-103 | 39.8 d | 0.498 | 50 | 25 |
| Ce-Pr-141 | 33.1 d | 0.142 | 14 | 7 |
| Co-57 | 270 d | 0.123 | 12 | 6 |
| Na-22 | 2.58 y | 0.510 | 51 | 25 |
| | | 1.277 | 128 | 64 |
| Mn-54 | 291 d | 0.840 | 84 | 42 |
| Co-60 | 5.24 y | 1.173 | 117 | 59 |
| | | 1.333 | 133 | 67 |
| Ba-133 | 7.2 y | 0.360 | 36 | 18 |

*Half-life of parent nuclide.

Routine Counting Procedure

(1) At the beginning of each workday, small cesium-137 and cobalt-60 standards (point sources) are counted for 10 minutes in a uniform geometry. From the location of the peaks, the energy alignment of the spectrometer is checked and adjusted, if necessary. A check on counting efficiency is obtained by comparing the counts in selected spectral regions to the usual counts for these sources. A plot of these daily counts as a function of date will indicate sudden degradation in counting efficiency as well as long-term changes in efficiency if such should occur. Radioactive decay of the standards must, of course, be taken into consideration. This daily procedure also checks out the operation of the complete spectrometer system, including the data read-out device.

(2) After checking the energy alignment and counting efficiency of the spectrometer system, a background count is made on each detector using a 3.5-liter sample of distilled water. The background should be counted for the same length of time as the samples are counted.

If the background is to be subtracted automatically by the spectrometer, the background should be stored in the spectrometer in the subtract mode. If background is not to be automatically subtracted, store background in the add mode, print out background, and erase from analyzer memory. Particular attention should be given to the background data to see that the backgrounds are normal and that there is no evidence of contamination of the detector or shield.

(3) Place the beaker containing the sample in position for counting. Count for the appropriate length of time. Usually a 50-minute counting time is used when the spectrometer is calibrated at 20 Kev/channel and a 100-minute counting time is used for a calibration of 10 Kev/channel.

Analysis of Spectral Data

The analysis of the spectral data obtained from the gamma spectrometer is an important and usually the most complicated aspect of the use of this instrument to measure radionuclide concentrations. If one is interested in only a qualitative determination of the radionuclides in a sample, i.e., identification of the gamma emitters, then the location of the peaks in the spectrum is of primary interest. If one desires to determine the amount of a particular nuclide in the sample, then the location of the peaks, the number of counts in the peaks, and the interference of one peak

with another all must be considered. A number of methods have been devised for analyzing gamma-spectral data. Two of these methods are in general use and are described below.

(1) Stripping Method: In this method the spectrum of each radionuclide is successively stripped from the composite spectrum of the sample. Beginning with the nuclide having the highest energy photopeak, the amount of this nuclide is first determined. The standard spectrum of this nuclide is then normalized to the high energy photopeak in the sample spectrum and the normalized standard spectrum is subtracted channel by channel from the sample spectrum. Having removed the spectrum of the highest energy gamma emitter from the composite spectrum, the next highest gamma-energy photopeak is selected, the amount of radioactivity determined, the standard spectrum for this radionuclide normalized to the sample spectrum, and the spectrum of this radionuclide stripped from the composite spectrum. This procedure is continued until all identifiable radionuclides present in the sample have been quantitized. It should be noted that the stripping method is most useful in the case when only a few gamma-emitting nuclides are present in the sample. Also, if there is direct interference between the photopeaks of different nuclides, the stripping method becomes somewhat inaccurate. A further disadvantage is that if a particular nuclide has more than one gamma, usually the highest energy gamma must be used as the normalizer and, in some cases, the photopeak from this gamma may be extremely low compared to other peaks in the spectrum. The errors associated with each successive "strip" are cumulative, causing the error to be great in the lower energy end of the spectrum.

If only one gamma-emitting radionuclide is present in the sample, the amount of the radionuclide may be determined directly from the net count (gross count-background) in the selected photopeak region. The same spectral region must be used in the sample spectrum as was used in the standard spectrum when counting efficiency was determined.

If only three or four gamma-emitting nuclides are present in the sample and if their photopeaks are separated in energy, a simplified stripping procedure may be used. The simplified procedure consists of subtracting the Compton spectrum of higher energy gammas from the photopeak of the lower energy gammas. In most cases the Compton spectra are appsoximately straight lines in the regions of the lower energy photopeaks, i.e., the counts do not vary greatly from one channel to the next. Thus, a straight line is drawn across the base of the photopeaks of interest and the counts above this baseline are taken as the counts due to the nuclide of interest. This procedure is

applied to a net spectrum, i.e., after the background has been subtracted. The following formula describes the procedure:

(Gross counts in spectral region of interest) - (Background counts) - (Baseline counts) = (Counts due to nuclide of interest)

(2) Matrix (Simultaneous Solution) Method: The matrix method is similar to the stripping method in that the interference of the spectrum of one nuclide with that of another is considered, but the interferences are subtracted simultaneously rather than successively as in the stripping method. Interference in regions of energy higher than the photopeak are considered as well as the interference in regions of lower energy. Also cases of direct interference of photopeaks can be handled by this method. The paper by Kahn, et al.,* gives a detailed description of the method.

In general, equations are set up describing the contribution of each radionuclide to the counts in a selected spectral region of interest. The number of equations corresponds to the number of nuclides. The contributions of each nuclide to a given spectral region are determined from the individual spectra of standard sources. The set of equations is then solved simultaneously. Such simultaneous solution is most easily performed by a computer when the number of nuclides (and hence the number of equations) is more than 2 or 3. The solutions of the set of equations are then new equations which give the count due to each nuclide of interest in terms of the observed counts in each spectral region and constant factors. In using these equations, one determines the net count (gross count-background) in each of the selected spectral regions of the sample spectrum, inserts these values into the equation, and computes the true count due to each nuclide. The counting efficiency factors are then used to obtain the amount of each radionuclide in the sample. It should be noted that inaccuracy may result if a nuclide is present in the sample but is not accounted for in the set of equations.

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*Kahn, Bernd, et al.: "Rapid Methods for Estimating Fission Product Concentrations in Milk", U.S. Department of Health, Education, and Welfare, Public Health Service Publication No. 999-R-2 (1963), p. 17.

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# APPENDIX A

## Preparation of Reagents

All reagents are analytical grade. Amounts of materials to be weighed correspond to the chemical formula listed. Some reagents may be obtained containing varying amounts of water of hydration; therefore, the usual corrections may be necessary for proper dilution. All anhydrous salts should be dried for 2 hours at 110° C before weighing.

### Carriers

The barium and yttrium carriers must be standardized. If recovery of strontium carrier is not determined flame-photometrically, the strontium carrier must also be standardized. All carriers should be checked for radioactive contamination by pipetting several 2-ml aliquots of each carrier solution into 2-in. planchets and counting each aliquot in the counting system appropriate to the carrier use. If the radioactive contamination of yttrium carrier exceeds 0.1 cpm/ml above background, the carrier solution should be purified by TBP extraction and reextraction. Restandardize after purification. Other carriers may have greater amounts of radioactive contamination, but should not exceed 0.2 cpm/ml above background under any circumstances.

1. Barium Carrier

| | |
|---|---|
| 20 mg $Ba^{+2}$/ml: | Dissolve 38.1 g of $Ba(NO_3)_2$ in 900 ml of distilled water. Add 1 ml of 16N $HNO_3$ and dilute to 1,000 ml with distilled water. |
| 5 mg $Ba^{+2}$/ml: | Dissolve 9.5 g of $Ba(NO_3)_2$ in 900 ml of distilled water. Add 1 ml of 16N $HNO_3$ and dilute to 1,000 ml with distilled water. |

2. Lead Carrier

| | |
|---|---|
| 100 mg $Pb^{+2}$/ml: | Dissolve 160.0 g of $Pb(NO_3)_2$ in 900 ml of distilled water. Add 1 ml of 16N $HNO_3$ and dilute to 1,000 ml with distilled water. |

Mixed Carrier

22 mg $Sr^{+2}$/ml and 5 mg/ml each of $Zn^{+2}$, $Fe^{+3}$, $Ni^{+2}$, and $Co^{+2}$:

Dissolve 53.2 g of $Sr(NO_3)_2$, 22.7 g of $Zn(NO_3)_2.6H_2O$, 24.2 g of $FeCl_3.6H_2O$, 24.4 g of $Ni(NO_3)_2.6H_2O$, and 24.3 g of $Co(NO_3)_2.6H_2O$ in 900 ml of distilled water. Add 1 ml of 16N $HNO_3$ and dilute to 1,000 ml with distilled water.

4. Mixed Rare Earth Carrier

10 mg/ml each of $Fe^{+3}$, $Ce^{+3}$, and $Zr^{+4}$:

Dissolve 48.4 g of $FeCl_3.6H_2O$, 31.0 g of $Ce(NO_3)_3.6H_2O$, and 35.5 g of $ZrOCl_2$ in 900 ml of distilled water. Add 1 ml of 16N $HNO_3$ and dilute to 1,000 ml with distilled water.

5. Strontium Carrier

20 mg $Sr^{+2}$/ml:

Dissolve 48.3 g of $Sr(NO_3)_2$ in 900 ml of deionized distilled water. Add 1 ml of 16N $HNO_3$ and dilute to 1,000 ml with deionized distilled water.

6. Strontium Standard Solution

0.4 mg $Sr^{+2}$/ml:

For flame-photometric determination of recovery of strontium carrier, dilute 1 ml of strontium carrier to 50 ml with deionized distilled water.

7. Yttrium Carrier

10 mg $Y^{+3}$/ml:

Heat (avoid boiling) 12.7 g of $Y_2O_3$ in 50 ml of 16N $HNO_3$ until dissolved. Transfer to 1-liter volumetric flask and dilute to 900 ml. Adjust to pH 2 with 15N $NH_4OH$ and dilute to 1,000 ml with distilled water.

## Reagents

### 1. Acetic Acid

18N: 99.5% $HC_2H_3O_2$, glacial, sp. gr. 1.06.

### 2. Aerosol OT

Dissolve approximately 3 g of Aerosol OT (solid pellets) in 250 ml of distilled water. Do not dilute. Reagent tends to favor *bacteriological growth so that* extended storage is not reasonable.

### 3. Ammonium Acetate Buffer

pH 5: Dissolve 153 g of ammonium acetate ($NH_4C_2H_3O_2$) in 900 ml of distilled water. Add 28.6 ml of glacial acetic acid (99.5%) to the solution. Dilute to 1,000 ml with distilled water.

### 4. Ammonium Carbonate

3N: Dissolve 144 g of $(NH_4)_2CO_3$ in 900 ml of distilled water and dilute to 1,000 ml with distilled water.

### 5. Ammonium Hydroxide

15N: 58% $NH_4OH$, sp. gr. 0.90.

6N: Dilute 400 ml of the 58% reagent to 1,000 ml with distilled water.

### 6. Ammonium Oxalate

1N: Dissolve 71 g of $(NH_4)_2C_2O_4.H_2O$ in 500 ml of hot distilled water and dilute to 1,000 ml with distilled water.

0.03N: Dissolve 2.1 g of $(NH_4)_2C_2O_4.H_2O$ in 900 ml of distilled water and dilute to 1,000 ml with distilled water.

7. Ammonium Sulfate

| | |
|---|---|
| 0.02N: | Dissolve 1.32 g of $(NH_4)_2SO_4$ in 900 ml of distilled water and dilute to 1,000 ml with distilled water. |

8. Anion Exchange Resin

| | |
|---|---|
| Dowex-1-X8 (50-100 mesh) ($Cl^-$ form): | Wash resin in column with 200 ml of 2N HCl, then 100 ml distilled water, each washing at a flow rate of 2 ml/min. Check pH of water effluent to insure complete removal of HCl. If pH is low, continue washing with distilled water until pH of eluent and effluent are essentially identical. |

9. Cation Exchange Resin

| | |
|---|---|
| Dowex-50W-X8 (50-100 mesh) ($H^+$ form) | |
| Dowex-50W-X8 (50-100 mesh) ($Na^+$ form): | Wash resin in column with 1,500 ml of 4N NaCl, then with 500 ml of distilled water, each washing at a flow rate of 10 ml/min. |

10. Citric Acid

| | |
|---|---|
| 3N: | Dissolve 210.1 g of $H_3C_6H_5O_7.H_2O$ in 900 ml of distilled water and dilute to 1,000 ml with distilled water. |

11. Citrate Solution

| | |
|---|---|
| 3N(pH 6.5): | Dissolve 210.1 g of citric acid ($H_3C_6H_5O_7.H_2O$) in 500 ml of distilled water. Adjust solution to pH 6.5 with approximately 220 ml of 12N NaOH (using pH meter). Dilute to 1,000 ml with distilled water. |

12. Ethylenediaminetetraacetate (disodium)

| | |
|---|---|
| 0.5N: | Dissolve 93.1 g of $Na_2C_{10}H_{14}O_8N_2.2H_2O$ in 900 ml of distilled water and dilute to 1,000 ml with distilled water. |

13. EDTA Wash Solution

| | |
|---|---|
| pH 10: | Dissolve 10 g of $Na_2C_{10}H_{14}O_8N_2.2H_2O$ in 950 ml of distilled water. Adjust solution to ≈ pH 10 with 6N NaOH and dilute to 1,000 ml with distilled water. |

14. Hydrochloric Acid

| | |
|---|---|
| 12N: | 37% HCl, sp. gr. 1.18. |
| 6N: | Dilute 500 ml of the 37% reagent to 1,000 ml with distilled water. |
| 3.5N: | Dilute 292 ml of the 37% reagent to 1,000 ml with deionized distilled water. |
| 2N: | Dilute 167 ml of the 37% reagent to 1,000 ml with distilled water. |
| 0.5N: | Dilute 41.7 ml of the 37% reagent to 1,000 ml with deionized distilled water. |

15. Hydrochloric Acid-Diethyl Ether Reagent

Mix 5 volumes of 12N HCl with 1 volume of anhydrous diethyl ether.

16. Methyl Orange Indicator

Dissolve 0.1 g of methyl orange in 100 ml of distilled water. Filter solution through Millipore #HA membrane filter (47-mm diameter) and discard membrane filter. Do not dilute solution further.

17. Methyl Purple Indicator

Obtained as solution from Fleisher Chemical Company, Benjamin Franklin Station, Washington 4, D.C. Solution is filtered through Millipore #HA membrane filter (47-mm diameter) and membrane filter is discarded. Do not dilute solution further.

18. Mixed Color Reagent

Dissolve 4.0 g of sodium metavanadate ($NaVO_3$) into a solution of 500 ml of 70% $HClO_4$ in 400 ml of deionized distilled water, heating to aid dissolution. Cool and dilute to 1,000 ml with deionized distilled water. Dissolve 80 g of sodium molybdate ($Na_2MoO_4.2H_2O$) in 900 ml deionized distilled water, heating to aid dissolution. Cool and dilute to 1,000 ml with deionized distilled water. Each solution is stable over extended periods of time. Prepare mixed color reagent immediately prior to use by combining equal volumes of the metavanadate solution and the molybdate solution.

19. Nitric Acid

| | |
|---|---|
| 90%: | $HNO_3$, fuming. |
| 16N: | 70% $HNO_3$, sp. gr. 1.413. |
| 14N: | Dilute 875 ml of the 70% reagent to 1,000 ml with distilled water. |
| 8N: | Dilute 500 ml with the 70% reagent to 1,000 ml with distilled water. |
| 6N: | Dilute 375 ml of the 70% reagent to 1,000 ml with distilled water. |
| 0.1N: | Dilute 6.25 ml of the 70% reagent to 1,000 ml with distilled water. |

**20. Oxalic Acid**

2N: Dissolve 126.1 g of $C_2H_2O_4.2H_2O$ in 900 ml of distilled water and dilute to 1,000 ml with distilled water.

1N: Dissolve 63.0 g of $C_2H_2O_4.2H_2O$ in 900 ml of distilled water and dilute to 1,000 ml with distilled water.

**21. pH 1 Wash Solution**

Dissolve 1.32 g of $(NH_4)_2SO_4$ in 950 ml of distilled water. Adjust solution to ≈ pH 1 with 1N $H_2SO_4$ and dilute to 1,000 ml with distilled water.

**22. Phosphate Standard Solution**

1 mg/ml $PO_4^{-3}$: Dissolve 1.4947 g of dry $Na_2HPO_4$ in 900 ml of deionized distilled water and dilute to 1,000 ml with deionized distilled water.

**23. Potassium Blank Solution**

3,500 ppm $Na^+$, 8,000 ppm $Ca^{++}$, and 120 ppm $Mg^{++}$: Dissolve 8.8990 g NaCl, 19.9800 g $CaCO_3$, and 4,700 g $MgCl_2$ in 900 ml 0.5N HCl and dilute to 1,000 ml with 0.5N HCl. All reagents should be analytically dried before weighing.

**24. Potassium Permanganate**

0.05N: Dissolve 1.6 g of $KMnO_4$ in 900 ml of distilled water. Heat to near boiling for 20-30 minutes and cool overnight. Filter through Whatman #1 filter paper into volumetric flask. Dilute to 1,000 ml with deionized distilled water.

**25. Potassium Standard Solution**

| | |
|---|---|
| 1,000 ppm $K^+$: | Dissolve 1.9069 g dry KCl in 900 ml of 0.5N HCl and dilute to 1,000 ml with 0.5N HCl. |

**26. Scintillation Mix**

| | |
|---|---|
| Dioxane: | Dissolve 4.0 g of PPO (2,5-diphenyl-oxazole), 0.05 g of POPOP [1,4-bis-2 (5-phenyl-oxozolyl)-benzene] and 120 g of naphthalene in 1,000 ml of p-dioxane.<br><br>All reagents should be spectro-quality grade if available. The p-dioxane must be freshly opened; if not, distillation is necessary.<br><br>This mix is stable for approximately 2 weeks. |

**27. Sodium Carbonate**

| | |
|---|---|
| 3N: | Dissolve 159 g of $Na_2CO_3$ in 900 ml of distilled water and dilute to 1,000 ml with distilled water. |
| 0.1N: | Dissolve 5.3 g of $Na_2CO_3$ in 900 ml of distilled water and dilute to 1,000 ml with distilled water. |

**28. Sodium Chloride**

| | |
|---|---|
| 4N: | Dissolve 234 g of NaCl in 900 ml of distilled water and dilute to 1,000 ml with distilled water. |

**29. Sodium Chromate**

| | |
|---|---|
| 3N: | Dissolve 243 g of $Na_2CrO_4$ in 900 ml of distilled water and dilute to 1,000 ml with distilled water. |

**30. Sodium Hydroxide**

| | |
|---|---|
| 6N: | Dissolve 240 g of NaOH in 900 ml of distilled water and dilute to 1,000 ml with distilled water. |

**31. Sodium Oxalate Standard**

Weigh out, in triplicate, 80 mg of dry $Na_2C_2O_4$ (NBS Primary Standard grade). Dissolve each in solutions of 1 ml of 12N HCl in 24 ml of distilled water.

**32. Strontium Standard Solution**

0.4 mg $Sr^{+2}$/ml: For flame-photometric determination of recovery of strontium carrier, dilute 1 ml of strontium carrier to 50 ml with deionized distilled water.

**33. Sulfuric Acid**

36N: 96% $H_2SO_4$, sp. gr. 1.84.

18N: Dilute 500 ml of the 96% reagent to 1,000 ml with distilled water.

1N: Dilute 278 ml of the 96% reagent to 1,000 ml with distilled water.

**34. Tributyl Phosphate (TBP)**

Preequilibrated: Prepare by adding 150 ml of distilled water and 30 ml of 3N $Na_2CO_3$ to 300 ml of TBP in a 1-liter separatory funnel. Equilibrate by shaking for 2-3 minutes. Allow phases to separate and discard lower aqueous phase. Add 150 ml of distilled water to the separatory funnel and equilibrate for 2-3 minutes. Allow phases to separate and discard lower aqueous phase. Add 150 ml of 14N $HNO_3$ to the separatory funnel and equilibrate for 5 minutes. Allow phases to separate and discard lower aqueous phase. Repeat this 14N $HNO_3$ treatment twice.

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## APPENDIX B

### Selected Radionuclide Decay and Ingrowth Factor Tables

Table 1: Y-90 Decay and Ingrowth Factors (0-72 hours)

Table 2: Y-90 Ingrowth Factors (0-27 days)

Table 3: Sr-89 Decay Factors (0.59.5 days)

Table 4: Ba-140 Decay Factors (0-50 days)

Table 5: La-140 Ingrowth Factors (0-72 hours)

Table 1. Y-90 Decay and Ingrowth Factors (0-72 Hours)

| t (hr) | $e^{-\lambda t}$ | $1-e^{-\lambda t}$ | t (hr) | $e^{-\lambda t}$ | $1-e^{-\lambda t}$ | t (hr) | $e^{-\lambda t}$ | $1-e^{-\lambda t}$ |
|---|---|---|---|---|---|---|---|---|
| 0.0 | 1.0000 | .0000 | 24.0 | .7717 | .2283 | 48.0 | .5955 | .4045 |
| 0.5 | .9946 | .0054 | 24.5 | .7676 | .2324 | 48.5 | .5923 | .4077 |
| 1.0 | .9893 | .0107 | 25.0 | .7634 | .2366 | 49.0 | .5891 | .4109 |
| 1.5 | .9839 | .0161 | 25.5 | .7593 | .2407 | 49.5 | .5860 | .4140 |
| 2.0 | .9786 | .0214 | 26.0 | .7552 | .2448 | 50.0 | .5828 | .4172 |
| 2.5 | .9734 | .0266 | 26.5 | .7512 | .2488 | 50.5 | .5797 | .4203 |
| 3.0 | .9681 | .0319 | 27.0 | .7471 | .2529 | 51.0 | .5766 | .4234 |
| 3.5 | .9629 | .0371 | 27.5 | .7431 | .2569 | 51.5 | .5735 | .4265 |
| 4.0 | .9577 | .0423 | 28.0 | .7391 | .2609 | 52.0 | .5704 | .4296 |
| 4.5 | .9526 | .0474 | 28.5 | .7351 | .2649 | 52.5 | .5673 | .4327 |
| 5.0 | .9474 | .0526 | 29.0 | .7311 | .2689 | 53.0 | .5642 | .4358 |
| 5.5 | .9423 | .0577 | 29.5 | .7272 | .2728 | 53.5 | .5612 | .4388 |
| 6.0 | .9373 | .0627 | 30.0 | .7233 | .2767 | 54.0 | .5582 | .4418 |
| 6.5 | .9322 | .0678 | 30.5 | .7194 | .2806 | 54.5 | .5552 | .4448 |
| 7.0 | .9272 | .0728 | 31.0 | .7155 | .2845 | 55.0 | .5522 | .4478 |
| 7.5 | .9222 | .0778 | 31.5 | .7117 | .2883 | 55.5 | .5492 | .4508 |
| 8.0 | .9172 | .0828 | 32.0 | .7078 | .2922 | 56.0 | .5462 | .4538 |
| 8.5 | .9123 | .0877 | 32.5 | .7040 | .2960 | 56.5 | .5433 | .4567 |
| 9.0 | .9074 | .0926 | 33.0 | .7002 | .2998 | 57.0 | .5404 | .4596 |
| 9.5 | .9025 | .0975 | 33.5 | .6965 | .3035 | 57.5 | .5375 | .4625 |
| 10.0 | .8976 | .1024 | 34.0 | .6927 | .3073 | 58.0 | .5346 | .4654 |
| 10.5 | .8928 | .1072 | 34.5 | .6890 | .3110 | 58.5 | .5317 | .4683 |
| 11.0 | .8880 | .1120 | 35.0 | .6853 | .3147 | 59.0 | .5288 | .4712 |
| 11.5 | .8832 | .1168 | 35.5 | .6816 | .3184 | 59.5 | .5260 | .4740 |
| 12.0 | .8785 | .1215 | 36.0 | .6779 | .3221 | 60.0 | .5232 | .4768 |
| 12.5 | .8737 | .1263 | 36.5 | .6743 | .3257 | 60.5 | .5203 | .4797 |
| 13.0 | .8690 | .1310 | 37.0 | .6706 | .3294 | 61.0 | .5175 | .4825 |
| 13.5 | .8644 | .1356 | 37.5 | .6670 | .3330 | 61.5 | .5148 | .4852 |
| 14.0 | .8597 | .1403 | 38.0 | .6634 | .3366 | 62.0 | .5120 | .4880 |
| 14.5 | .8551 | .1449 | 38.5 | .6599 | .3401 | 62.5 | .5092 | .4908 |
| 15.0 | .8505 | .1495 | 39.0 | .6563 | .3437 | 63.0 | .5065 | .4935 |
| 15.5 | .8459 | .1541 | 39.5 | .6528 | .3472 | 63.5 | .5038 | .4962 |
| 16.0 | .8413 | .1587 | 40.0 | .6493 | .3507 | 64.0 | .5010 | .4990 |
| 16.5 | .8368 | .1632 | 40.5 | .6458 | .3542 | 64.5 | .4983 | .5017 |
| 17.0 | .8323 | .1677 | 41.0 | .6423 | .3577 | 65.0 | .4957 | .5043 |
| 17.5 | .8278 | .1722 | 41.5 | .6388 | .3612 | 65.5 | .4930 | .5070 |
| 18.0 | .8234 | .1766 | 42.0 | .6354 | .3646 | 66.0 | .4903 | .5097 |
| 18.5 | .8189 | .1811 | 42.5 | .6320 | .3680 | 66.5 | .4877 | .5123 |
| 19.0 | .8145 | .1855 | 43.0 | .6286 | .3714 | 67.0 | .4851 | .5149 |
| 19.5 | .8101 | .1899 | 43.5 | .6252 | .3748 | 67.5 | .4825 | .5175 |
| 20.0 | .8058 | .1942 | 44.0 | .6219 | .3781 | 68.0 | .4799 | .5201 |
| 20.5 | .8014 | .1986 | 44.5 | .6185 | .3815 | 68.5 | .4773 | .5227 |
| 21.0 | .7971 | .2029 | 45.0 | .6151 | .3849 | 69.0 | .4747 | .5253 |
| 21.5 | .7928 | .2072 | 45.5 | .6118 | .3882 | 69.5 | .4722 | .5278 |
| 22.0 | .7885 | .2115 | 46.0 | .6085 | .3915 | 70.0 | .4696 | .5304 |
| 22.5 | .7843 | .2157 | 46.5 | .6053 | .3947 | 70.5 | .4671 | .5329 |
| 23.0 | .7801 | .2199 | 47.0 | .6020 | .3980 | 71.0 | .4646 | .5354 |
| 23.5 | .7759 | .2241 | 47.5 | .5988 | .4012 | 71.5 | .4621 | .5379 |
| | | | | | | 72.0 | .4596 | .5404 |

Table 2. Y-90 Ingrowth Factors (0-27 Days)

| t (days) | $1-e^{-\lambda t}$ | t (days) | $1-3^{-\lambda t}$ | t (days) | $1-e^{-\lambda t}$ |
|---|---|---|---|---|---|
| 0.00 | .0000 | 9.00 | .9029 | 18.00 | .9906 |
| 0.25 | .0627 | 9.25 | .9090 | 18.25 | .9912 |
| 0.50 | .1215 | 9.50 | .9147 | 18.50 | .9917 |
| 0.75 | .1766 | 9.75 | .9201 | 18.75 | .9922 |
| 1.00 | .2283 | 10.00 | .9251 | 19.00 | .9927 |
| 1.25 | .2767 | 10.25 | .9298 | 19.25 | .9932 |
| 1.50 | .3221 | 10.50 | .9342 | 19.50 | .9936 |
| 1.75 | .3646 | 10.75 | .9384 | 19.75 | .9940 |
| 2.00 | .4045 | 11.00 | .9422 | 20.00 | .9944 |
| 2.25 | .4418 | 11.25 | .9458 | 20.25 | .9948 |
| 2.50 | .4768 | 11.50 | .9492 | 20.50 | .9951 |
| 2.75 | .5097 | 11.75 | .9524 | 20.75 | .9954 |
| 3.00 | .5404 | 12.00 | .9554 | 21.00 | .9957 |
| 3.25 | .5692 | 12.25 | .9582 | 21.25 | .9959 |
| 3.50 | .5963 | 12.50 | .9608 | 21.50 | .9962 |
| 3.75 | .6216 | 12.75 | .9633 | 21.75 | .9964 |
| 4.00 | .6453 | 13.00 | .9656 | 22.00 | .9967 |
| 3.25 | .6676 | 13.25 | .9678 | 22.25 | .9969 |
| 4.50 | .6884 | 13.50 | .9697 | 22.50 | .9971 |
| 4.75 | .7080 | 13.75 | .9716 | 22.75 | .9973 |
| 5.00 | .7263 | 14.00 | .9734 | 23.00 | .9974 |
| 5.25 | .7435 | 14.25 | .9751 | 23.25 | .9976 |
| 5.50 | .7596 | 14.50 | .9766 | 23.50 | .9977 |
| 5.75 | .7746 | 14.75 | .9781 | 23.75 | .9979 |
| 6.00 | .7888 | 15.00 | .9795 | 24.00 | .9980 |
| 6.25 | .8020 | 15.25 | .9808 | 24.25 | .9981 |
| 6.50 | .8145 | 15.50 | .9820 | 24.50 | .9982 |
| 6.75 | .8261 | 15.75 | .9831 | 24.75 | .9984 |
| 7.00 | .8370 | 16.00 | .9842 | 25.00 | .9985 |
| 7.25 | .8472 | 16.25 | .9852 | 25.25 | .9986 |
| 7.50 | .8568 | 16.50 | .9861 | 25.50 | .9987 |
| 7.75 | .8658 | 16.75 | .9870 | 25.75 | .9987 |
| 8.00 | .8742 | 17.00 | .9878 | 26.00 | .9988 |
| 8.25 | .8820 | 17.25 | .9886 | 26.25 | .9989 |
| 8.50 | .8896 | 17.50 | .9893 | 26.50 | .9990 |
| 8.75 | .8964 | 17.75 | .9900 | 26.75 | .9990 |
| | | | | 27.00 | .9991 |

Table 3. Sr-89 Decay Factors (0-59.5 Days)

($t_{1/2}$ = 51 days)

| t (days) | $e^{-\lambda t}$ | t (days) | $e^{-\lambda t}$ | t (days) | $e^{-\lambda t}$ |
|---|---|---|---|---|---|
| 0.0 | 1.0000 | 20.0 | .7620 | 40.0 | .5808 |
| 0.5 | .9932 | 20.5 | .7569 | 40.5 | .5769 |
| 1.0 | .9865 | 21.0 | .7518 | 41.0 | .5730 |
| 1.5 | .9798 | 21.5 | .7468 | 41.5 | .5690 |
| 2.0 | .9732 | 22.0 | .7416 | 42.0 | .5652 |
| 2.5 | .9668 | 22.5 | .7366 | 42.5 | .5613 |
| 3.0 | .9601 | 23.0 | .7317 | 43.0 | .5575 |
| 3.5 | .9536 | 23.5 | .7267 | 43.5 | .5539 |
| 4.0 | .9471 | 24.0 | .7218 | 44.0 | .5500 |
| 4.5 | .9407 | 24.5 | .7169 | 44.5 | .5462 |
| 5.0 | .9344 | 25.0 | .7120 | 45.0 | .5427 |
| 5.5 | .9280 | 25.5 | .7072 | 45.5 | .5380 |
| 6.0 | .9217 | 26.0 | .7023 | 46.0 | .5352 |
| 6.5 | .9155 | 26.5 | .6977 | 46.5 | .5318 |
| 7.0 | .9093 | 27.0 | .6930 | 47.0 | .5280 |
| 7.5 | .9031 | 27.5 | .6882 | 47.5 | .5245 |
| 8.0 | .8970 | 28.0 | .6836 | 48.0 | .5210 |
| 8.5 | .8909 | 28.5 | .6790 | 48.5 | .5175 |
| 9.0 | .8849 | 29.0 | .6742 | 49.0 | .5140 |
| 9.5 | .8789 | 29.5 | .6699 | 49.5 | .5105 |
| 10.0 | .8729 | 30.0 | .6651 | 50.0 | .5070 |
| 10.5 | .8670 | 30.5 | .6608 | 50.5 | .5035 |
| 11.0 | .8612 | 31.0 | .6562 | 51.0 | .5000 |
| 11.5 | .8553 | 31.5 | .6519 | 51.5 | .4967 |
| 12.0 | .8495 | 32.0 | .6473 | 52.0 | .4933 |
| 12.5 | .8438 | 32.5 | .6430 | 52.5 | .4900 |
| 13.0 | .8381 | 33.0 | .6388 | 53.0 | .4868 |
| 13.5 | .8324 | 33.5 | .6342 | 53.5 | .4834 |
| 14.0 | .8268 | 34.0 | .6300 | 54.0 | .4801 |
| 14.5 | .8212 | 34.5 | .6259 | 54.5 | .4769 |
| 15.0 | .8156 | 35.0 | .6215 | 55.0 | .4734 |
| 15.5 | .8101 | 35.5 | .6172 | 55.5 | .4702 |
| 16.0 | .8046 | 36.0 | .6131 | 56.0 | .4671 |
| 16.5 | .7992 | 36.5 | .6090 | 56.5 | .4640 |
| 17.0 | .7938 | 37.0 | .6050 | 57.0 | .4608 |
| 17.5 | .7883 | 37.5 | .6009 | 57.5 | .4578 |
| 18.0 | .7881 | 38.0 | .5968 | 58.0 | .4547 |
| 18.5 | .7778 | 38.5 | .5928 | 58.5 | .4513 |
| 19.0 | .7725 | 39.0 | .5888 | 59.0 | .4484 |
| 19.5 | .7672 | 39.5 | .5848 | 59.5 | .4454 |

Table 4. Ba-140 Decay Factors (0-50 Days)

(t 1/2 = 12.8 days)

| t (days) | e-λt | t (days) | e-λt | t (days) | e-λt |
|---|---|---|---|---|---|
| 0.0 | 1.0000 | 17.0 | .3983 | 34.0 | .1587 |
| 0.5 | .9733 | 17.5 | .3878 | 34.5 | .1544 |
| 1.0 | .9473 | 18.0 | .3773 | 35.0 | .1502 |
| 1.5 | .9219 | 18.5 | .3671 | 35.5 | .1462 |
| 2.0 | .8974 | 19.0 | .3572 | 36.0 | .1423 |
| 2.5 | .8734 | 19.5 | .3478 | 36.5 | .1386 |
| 3.0 | .8501 | 20.0 | .3385 | 37.0 | .1349 |
| 3.5 | .8273 | 20.5 | .3293 | 37.5 | .1312 |
| 4.0 | .8053 | 21.0 | .3206 | 38.0 | .1277 |
| 4.5 | .7838 | 21.5 | .3120 | 38.5 | .1243 |
| 5.0 | .7628 | 22.0 | .3038 | 39.0 | .1210 |
| 5.5 | .7425 | 22.5 | .2957 | 39.5 | .1178 |
| 6.0 | .7226 | 23.0 | .2878 | 40.0 | .1147 |
| 6.5 | .7033 | 23.5 | .2800 | 40.5 | .1116 |
| 7.0 | .6845 | 24.0 | .2725 | 41.0 | .1087 |
| 7.5 | .6662 | 24.5 | .2652 | 41.5 | .1058 |
| 8.0 | .6485 | 25.0 | .2581 | 42.0 | .1029 |
| 8.5 | .6311 | 25.5 | .2513 | 42.5 | .1001 |
| 9.0 | .6142 | 26.0 | .2447 | 43.0 | .0975 |
| 9.5 | .5980 | 26.5 | .2381 | 43.5 | .0949 |
| 10.0 | .5819 | 27.0 | .2318 | 44.0 | .0924 |
| 10.5 | .5663 | 27.5 | .2256 | 44.5 | .0900 |
| 11.0 | .5512 | 28.0 | .2195 | 45.0 | .0876 |
| 11.5 | .5365 | 28.5 | .2137 | 45.5 | .0853 |
| 12.0 | .5221 | 29.0 | .2079 | 46.0 | .0830 |
| 12.5 | .5082 | 29.5 | .2023 | 46.5 | .0807 |
| 13.0 | .4948 | 30.0 | .1970 | 47.0 | .0786 |
| 13.5 | .4815 | 30.5 | .1918 | 47.5 | .0765 |
| 14.0 | .4816 | 31.0 | .1866 | 48.0 | .0744 |
| 14.5 | .4531 | 31.5 | .1811 | 48.5 | .0725 |
| 15.0 | .4440 | 32.0 | .1765 | 49.6 | .0766 |
| 15.5 | .4320 | 32.5 | .1721 | 49.5 | .0687 |
| 16.0 | .4203 | 33.0 | .1675 | 50.0 | .0668 |
| 16.5 | .4091 | 33.5 | .1630 | | |

Table 5. La-140 Ingrowth Factors (0-72 Hours)

$(1-e^{-.015t})$

| t (hr) | $1-e^{-\lambda t}$ | t (hr) | $1-e^{-\lambda t}$ | t (hr) | $1-e^{-\lambda t}$ |
|---|---|---|---|---|---|
| 0.0 | .0000 | 24.0 | .3023 | 48.0 | .5133 |
| 0.5 | .0075 | 24.5 | .3075 | 48.5 | .5170 |
| 1.0 | .0149 | 25.0 | .3127 | 49.0 | .5207 |
| 1.5 | .0222 | 25.5 | .3179 | 49.5 | .5242 |
| 2.0 | .0295 | 26.0 | .3231 | 50.0 | .5277 |
| 2.5 | .0368 | 26.5 | .3281 | 50.5 | .5313 |
| 3.0 | .0440 | 27.0 | .3331 | 51.0 | .5347 |
| 3.5 | .0511 | 27.5 | .3381 | 51.5 | .5382 |
| 4.0 | .0582 | 28.0 | .3431 | 52.0 | .5417 |
| 4.5 | .0652 | 28.5 | .3480 | 52.5 | .5451 |
| 5.0 | .0722 | 29.0 | .3529 | 53.0 | .5485 |
| 5.5 | .0792 | 29.5 | .3578 | 53.5 | .5519 |
| 6.0 | .0860 | 30.0 | .3626 | 54.0 | .5552 |
| 6.5 | .0928 | 30.5 | .3673 | 54.5 | .5585 |
| 7.0 | .0996 | 31.0 | .3720 | 55.0 | .5618 |
| 7.5 | .1064 | 31.5 | .3767 | 55.5 | .5651 |
| 8.0 | .1131 | 32.0 | .3813 | 56.0 | .5684 |
| 8.5 | .1197 | 32.5 | .3859 | 56.5 | .5717 |
| 9.0 | .1263 | 33.0 | .3905 | 57.0 | .5749 |
| 9.5 | .1328 | 33.5 | .3951 | 57.5 | .5780 |
| 10.0 | .1393 | 34.0 | .3996 | 58.0 | .5811 |
| 10.5 | .1458 | 34.5 | .4041 | 58.5 | .5842 |
| 11.0 | .1522 | 35.0 | .4085 | 59.0 | .5873 |
| 11.5 | .1585 | 35.5 | .4129 | 59.5 | .5904 |
| 12.0 | .1648 | 36.0 | .4173 | 60.0 | .5935 |
| 12.5 | .1711 | 36.5 | .4217 | 60.5 | .5966 |
| 13.0 | .1773 | 37.0 | .4260 | 61.0 | .5996 |
| 13.5 | .1834 | 37.5 | .4303 | 61.5 | .6026 |
| 14.0 | .1895 | 38.0 | .4346 | 62.0 | .6056 |
| 14.5 | .1956 | 38.5 | .4388 | 62.5 | .6086 |
| 15.0 | .2016 | 39.0 | .4430 | 63.0 | .6115 |
| 15.5 | .2076 | 39.5 | .4472 | 63.5 | .6144 |
| 16.0 | .2135 | 40.0 | .4513 | 64.0 | .6172 |
| 16.5 | .2193 | 40.5 | .4554 | 64.5 | .6200 |
| 17.0 | .2251 | 41.0 | .4595 | 65.0 | .6229 |
| 17.5 | .2309 | 41.5 | .4636 | 65.6 | .6258 |
| 18.0 | .2367 | 42.0 | .4677 | 66.0 | .6286 |
| 18.5 | .2424 | 42.5 | .4716 | 66.5 | .6313 |
| 19.0 | .2481 | 43.0 | .4755 | 67.0 | .6340 |
| 19.5 | .2538 | 43.5 | .4794 | 67.5 | .6367 |
| 20.0 | .2593 | 44.0 | .4833 | 68.0 | .6394 |
| 20.5 | .2648 | 44.5 | .4871 | 68.5 | .6421 |
| 21.0 | .2703 | 45.0 | .4909 | 69.0 | .6448 |
| 21.5 | .2758 | 45.5 | .4947 | 69.5 | .6475 |
| 22.0 | .2811 | 46.0 | .4985 | 70.0 | .6502 |
| 22.5 | .2866 | 46.5 | .5022 | 70.5 | .6528 |
| 23.0 | .2919 | 47.0 | .5059 | 71.0 | .6554 |
| 23.5 | .2971 | 47.5 | .5096 | 71.5 | .6579 |
| | | | | 72.0 | .6604 |